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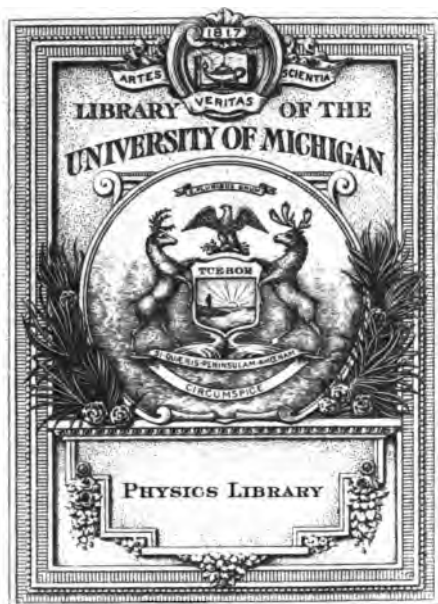
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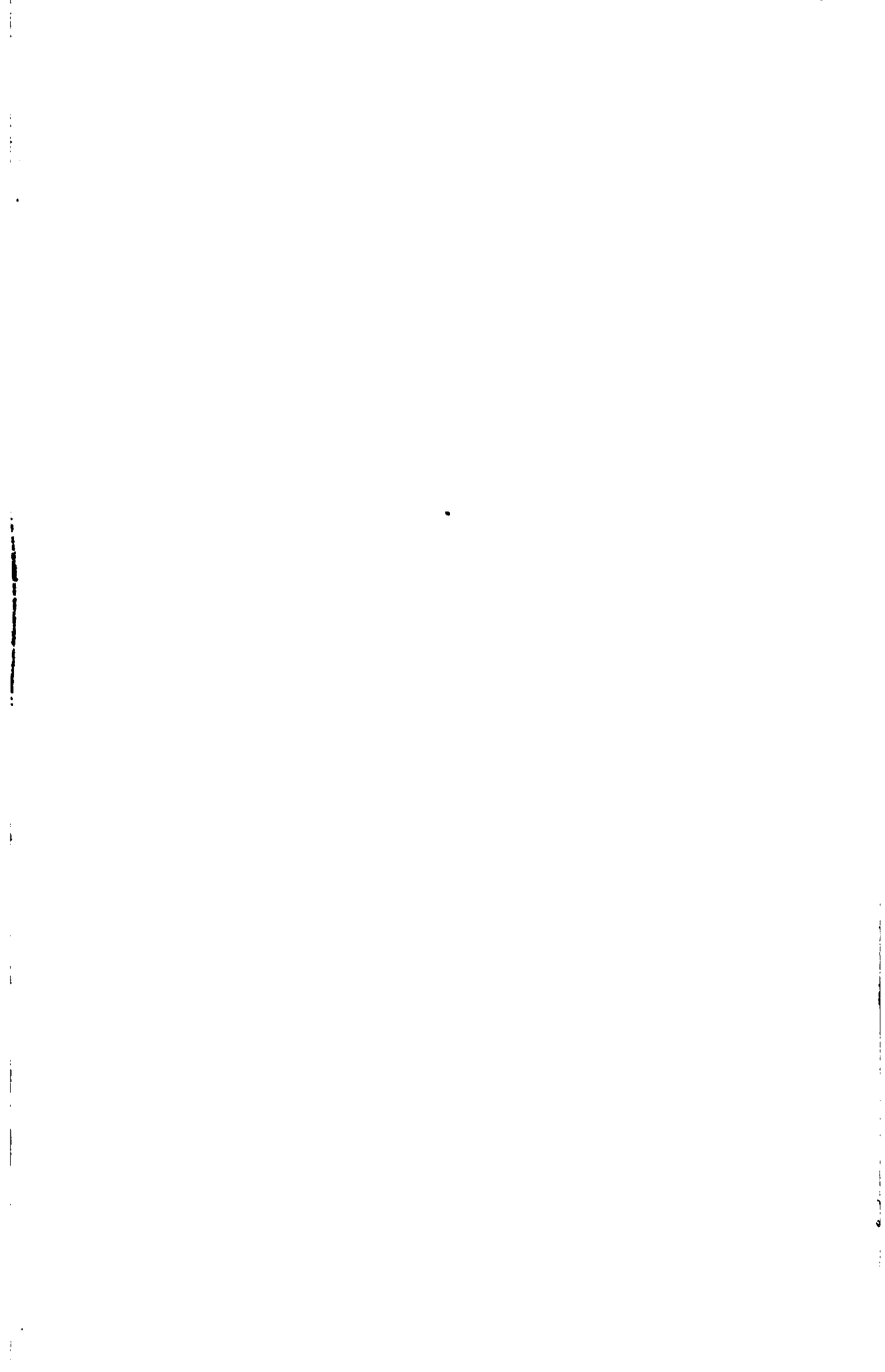
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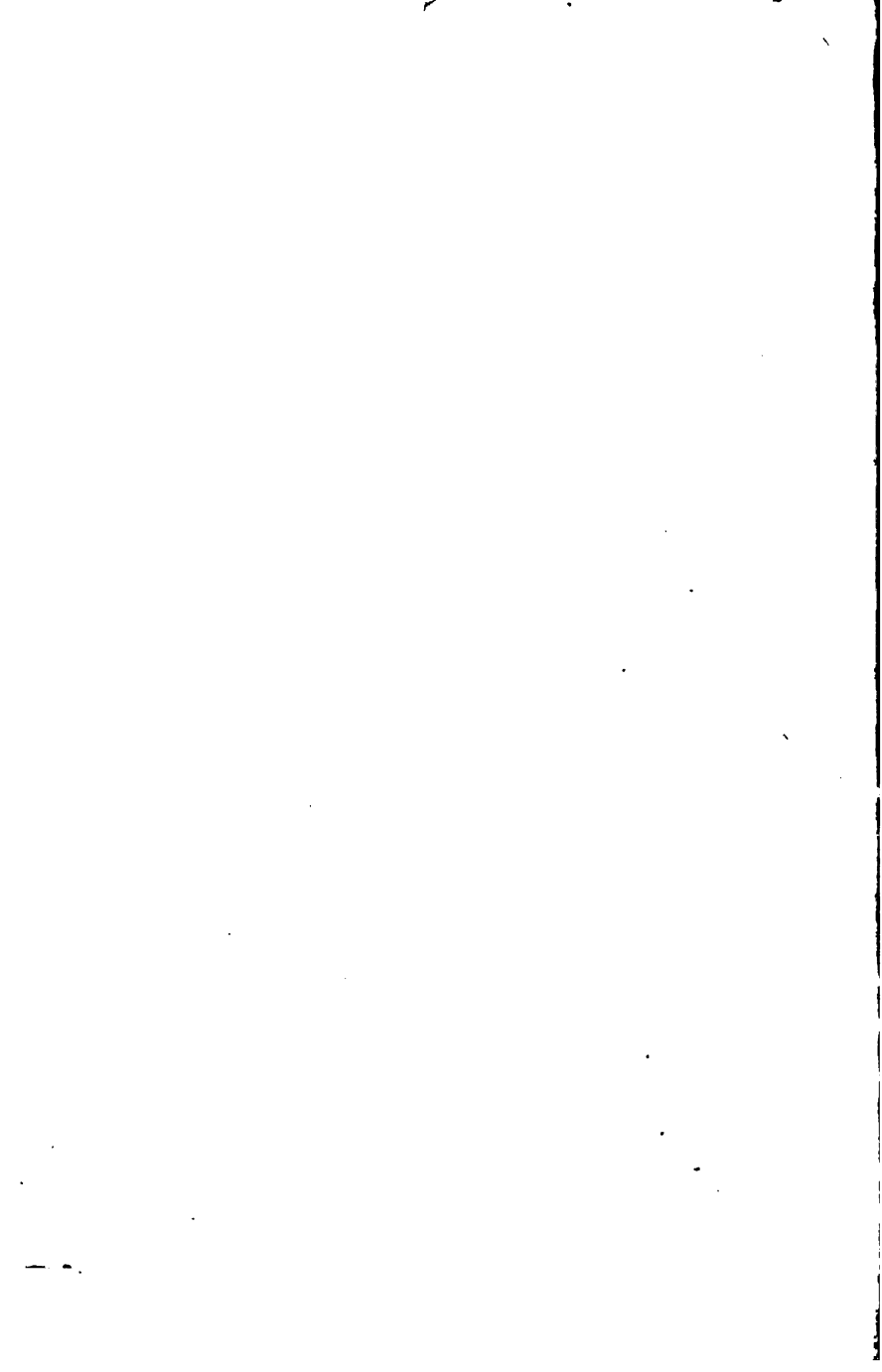
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# THE RADIOACTIVE SUBSTANCES

THEIR PROPERTIES AND BEHAVIOUR

BY

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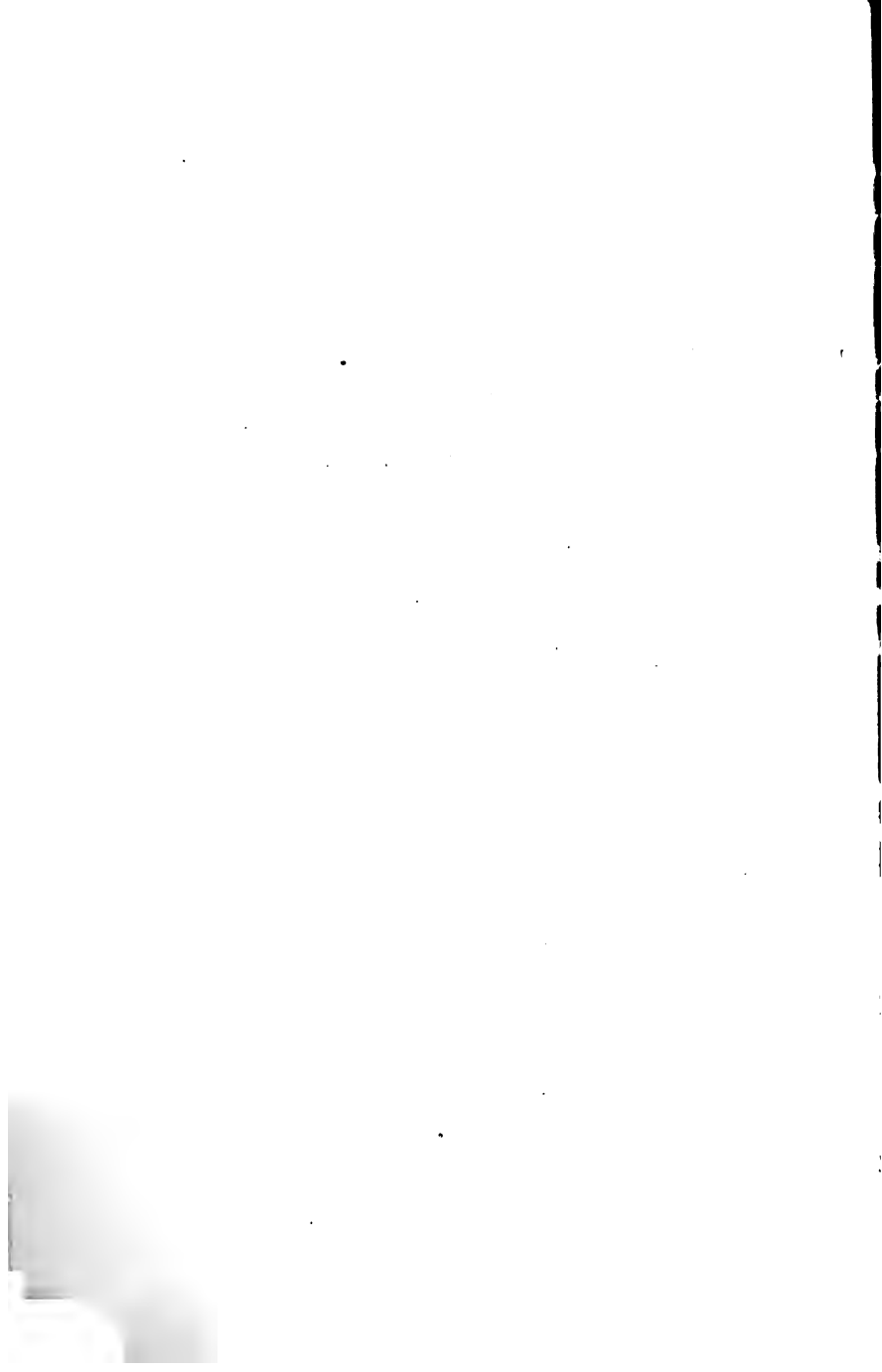
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1908

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## PREFACE

**I**N writing this volume an attempt has been made to present the chief phenomena and theories relating to radio-activity in a concise and simple form. It is unnecessary to point out the difficulty of doing this in a subject in which the results of new and important investigations are being published almost daily. But it is just this circumstance which has prompted the author to undertake the task of writing another book on a subject which has already been competently dealt with in an elementary way in other works.

It is manifestly impossible for those not specially engaged on the investigation of radioactive phenomena to keep pace with the literature of the subject, and it is hoped that to those who wish to make themselves acquainted with the general state of our knowledge of radio-activity without going into

undue detail, this book may be of use. The work therefore makes no claim to giving a complete account of all that is known about radio-activity, and those investigations and theories only have been selected for discussion which throw most light on the general phenomena of radio-activity.

Although only a few years have elapsed since the discovery of radio-activity by Becquerel, such has been the diligence with which the study of the properties of radioactive bodies has been pursued and such the skill with which the various phenomena have been co-ordinated, that the processes taking place during radioactive changes are already very fully understood. If we consider the influence which the discoveries in radio-activity are now exerting on modern views, both in physics and in chemistry, it is unnecessary to insist on the necessity of an understanding of this subject by students of both these sciences. It is hoped that to such students this book may serve as an introduction to the study of radio-activity. Only an elementary knowledge of either physics or



chemistry is necessary for an understanding of the matter dealt with, and the use of mathematics has been avoided except in a few special cases where the introduction of simple mathematical calculations seemed to tend to greater clearness. Thus in dealing with the disintegration theory the use of mathematics has been almost entirely avoided. This subject is somewhat difficult to explain without the use of mathematical analysis, but an attempt has been made even in this case to make the matter clear by means of a water analogy used to explain the transformation of radium A into radium B and radium C. Possibly this new method of presenting the subject may prove of assistance to the beginner.

References have frequently been given in footnotes to the sources from which information has been derived, but the author has consulted the treatise of Professor Rutherford on Radioactivity on so many points that it is only fitting that he should here express the obligation which he feels to that work. The special thanks of the author are due to

his colleague, Mr. S. Russ, of the Physical Department of the University of Manchester, for reading and correcting the manuscript and for many valuable suggestions and criticisms, and to Miss White for help in correcting the proof sheets.

W. M.

MANCHESTER, 11 *March*, 1908.

# THE RADIOACTIVE SUBSTANCES

## CHAPTER I

### INTRODUCTION

THE phenomena associated with the passage of electricity through gases have for some time past attracted the attention of scientific investigators. Not only are the appearances of the discharge, particularly at low pressures, of themselves of such a nature as to stimulate inquiry, but it has long been felt that, complicated as these manifestations appear at first sight, they are in many ways the most fundamental of all electrical phenomena. Researches of recent years have amply justified this belief, and have thrown light not only upon the nature of electricity but also upon the constitution of matter. But the discovery that certain substances spontaneously emit

radiations analogous to those now known to be produced during the passage of electricity through rarefied gases has surpassed the expectations even of the most sanguine. Such substances are said to be *radioactive*, and their discovery has necessitated a revision of many of our conceptions both in physics and chemistry.

The discovery of these radioactive substances was due to the fact that they emit radiations having certain peculiar properties, one of the most remarkable of which is to render gases through which they pass feeble conductors of electricity. It will therefore be necessary, before proceeding to a study of the phenomena of radio-activity, to study some of the most important facts connected with the conduction of electricity through gases.

#### CONDUCTIVITY OF GASES IN THE NORMAL STATE

It has long been known that a charged conductor suspended in air does not hold its charge for an indefinite time however great be the care with which it is insulated. As long ago as 1785, Coulomb concluded, as a result

of experiments on this point, that, allowing for the leakage over the insulation supporting a charged conductor suspended in air, there was still a loss of electricity which could not be accounted for by this cause. It seemed therefore that a current of electricity **must** have travelled through the gas surrounding the conductor. Although the subject has since received the attention of many physicists, the question was not finally settled until 1900, when Professor Geitel and Mr. C. T. R. Wilson independently published the results of investigations which left no doubt as to the property possessed by air of conducting electricity.

The method of investigation consisted in charging a well-insulated gold-leaf electroscope constructed in such a way that electricity could leak on to the leaves over the insulation if faulty, but that the reverse could not take place. Thus the effect of faulty insulation in such an experiment would have been to increase the charge originally communicated to the leaves of the electroscope, and any *diminution* could only be attributed

to a current passing through the gas to the walls of the electroscope case ; the current though exceedingly small is, however, within the means of refined measurement.

The conductivity of gases under ordinary circumstances is very slight, but it can be greatly increased in several ways.

#### METHODS OF INCREASING THE CONDUCTIVITY OF GASES

If, for instance, the gases from the neighbourhood of an ordinary gas-burner are withdrawn and examined, it is found that electricity is transmitted through them far more easily than through ordinary air. Again, the air in the neighbourhood of a solid heated to incandescence often conducts electricity quite readily, indeed frequently to such an extent that it has not been necessary to employ specially refined methods of measurements to detect, passing through the air, currents which are so large that they can be measured by means of a fairly sensitive galvanometer. As a further example of a similar effect on the conducting power of

gases may be mentioned the action of light of such short wave-length as to be incapable of detection by the eye. Such light, which is known as ultra-violet radiation, when it impinges on certain metallic plates causes the air in the neighbourhood of the plates to conduct electricity to a marked extent.

The properties exhibited by gases which have been rendered conducting by the above agencies have been carefully and exhaustively studied by many investigators, but the consideration of these phenomena, interesting though they may be, is beyond the scope of this work and must here be passed over. It is, however, important that it should be realized at the outset that gases may be converted into conductors of electricity by a variety of agencies.

Now there are certain other methods, besides those already mentioned, of throwing a gas into a conducting state which are particularly important from our present point of view. When a discharge of electricity is transmitted through a tube containing a rarefied gas, radiations may under suitable con-

ditions be obtained outside the discharge tube, which are capable of producing conductivity of electricity in the gases through which they pass. Of such a nature are, for instance, the radiations discovered by Röntgen as a result of their photographic action; the properties of these rays will be considered later. Again, there are certain substances, said to be "radioactive" and endowed with similar properties, which appear to continually emit radiations capable of rendering gases in their neighbourhood conductors. The properties and nature of these radiations will also be fully described later, but it will first be necessary for the better understanding of the subject briefly to consider at least some of the most important phenomena exhibited by gases when rendered conducting by the passage of radiations through them.

When a gas has been rendered a conductor of electricity, it retains this property only for a time, the length of which depends upon the manner in which it is treated. The property is one which has been acquired by the gas and which can be removed from it. If, for



instance, the gas be filtered through cotton wool, the conductivity is lost; or again, if it be passed through a strong electric field, it is restored to its normal condition. If, however, the gas is merely rapidly transferred from one place to another, it is found to have at least partially retained its conducting power. From these facts it would appear that the conductivity of the gas is due to the presence in it of certain material particles which can be removed by mechanical means, and further, that these particles are charged with electricity since they are removed by an electric field. These carriers are known as *ions*, and the gas is said to be *ionized* when it contains these carriers. The ions may be charged either with positive or negative electricity, and will therefore move towards the positive or negative pole when exposed to an electric field, according to the sign of the electricity which they carry.

#### LAW GOVERNING THE CONDUCTION OF ELECTRICITY THROUGH GASES

One of the most striking facts about the conduction of electricity through a gas is

that, except for very small electromotive forces, the current does not obey Ohm's Law, so that in this way a gas behaves differently from metallic and electrolytic conductors. The relation which exists between the electromotive force and the current has been very carefully worked out by Professors J. J. Thomson and Rutherford. The general features of the relation are shown in Fig. 1. As the field is increased the current rises at first proportionately to the electromotive force, and therefore in this region Ohm's Law is true. As the current is further increased the current rises less rapidly than the electromotive force, ultimately becoming nearly constant, and when this state is reached the current is said to be "saturated." This term has been borrowed from the somewhat analogous behaviour of iron magnetized by induction, for as the magnetizing force is increased the magnetic induction reaches a maximum, and the iron is said to be saturated with magnetism. The term is perhaps not a very happy one, as the saturation current depends on the degree of ionization and is therefore not an unalterable

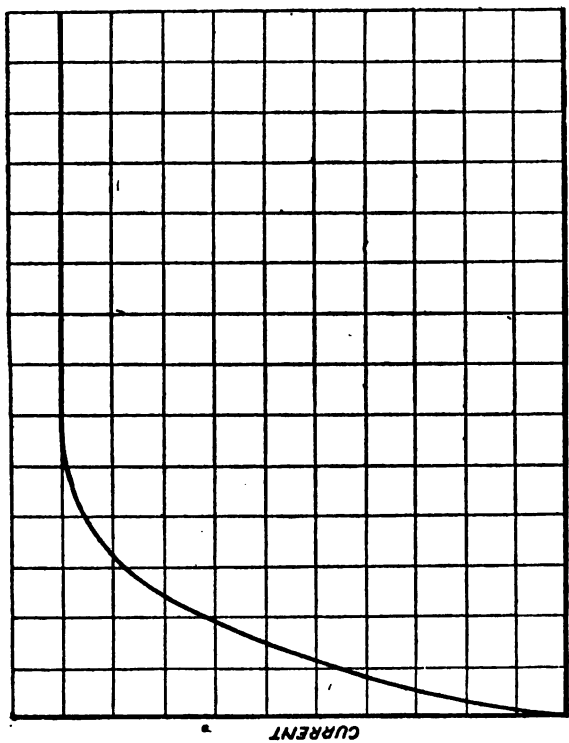
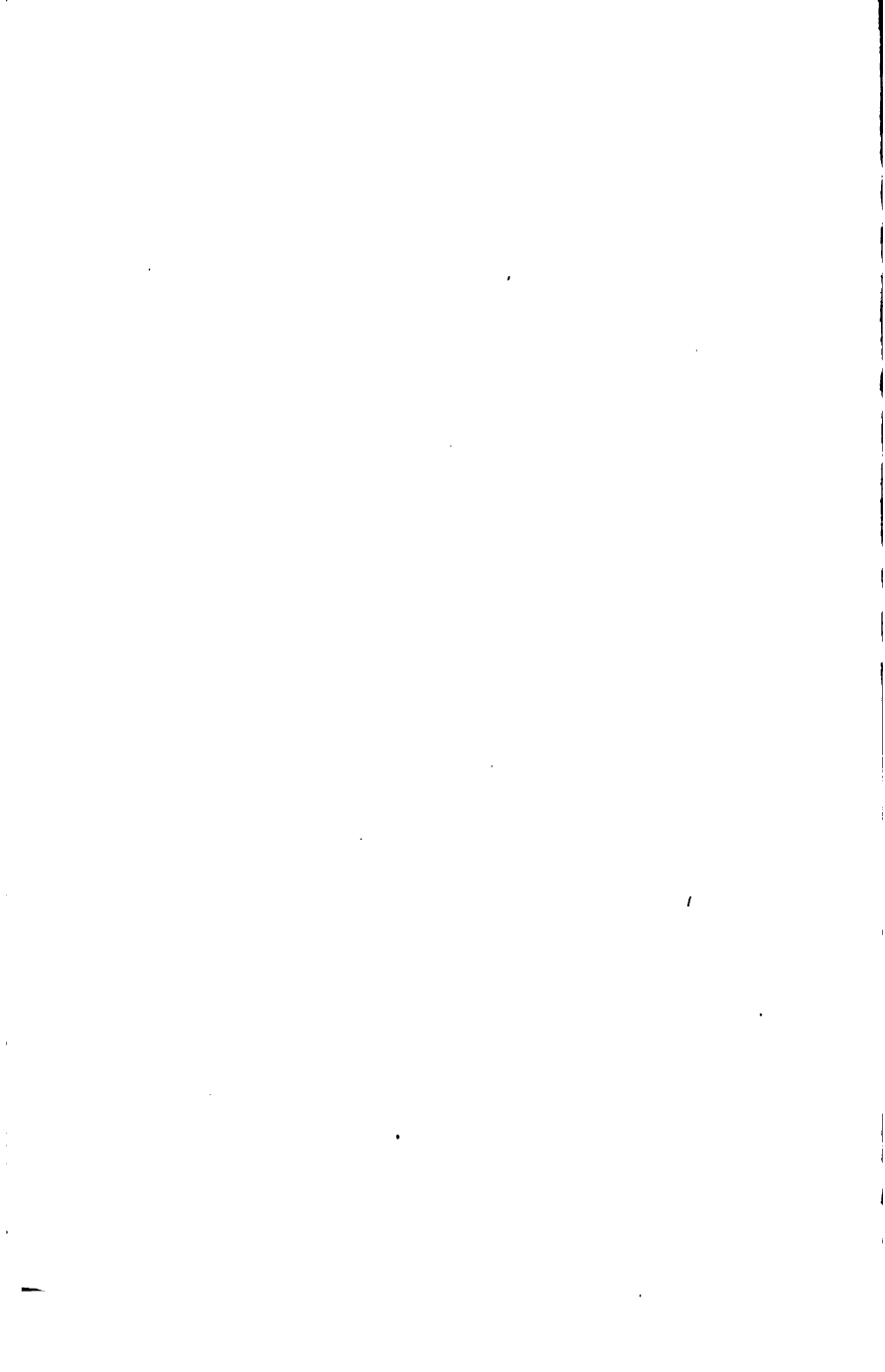


FIG. 1.—Curve showing relation of current in gases to E.M.F.



property of gas; it has, however, come into general use and must therefore be retained.

Let us consider a little more closely what is the physical meaning of the saturation current. We have seen that the current which flows through the gas is due to charged ions which are removed by the electric field. Now these ions may be considered as being formed by the ionizing rays at a constant rate, and a certain proportion of them is being removed by the current in every second. Now as the electromotive force is increased, more and more ions will be removed per second, and consequently the current through the gas will increase. When, however, the rate at which the ions are being produced is equal to the rate at which they are being removed by the electric field, an increase in the electromotive force can produce no further effect on the current, and it is this state of affairs which corresponds to the saturation current. Although it is a matter of no great difficulty to approximate to saturation (a potential gradient of 100 volts per cm. is usually sufficient), it re-

quires a very large potential difference to reach a true saturation current, and it would seem that a small proportion of the ions are so heavy that it requires a strong electric force to remove these before sufficient time has elapsed for the ions carrying opposite signs to come into contact with each other and to neutralize each other's charge. This process which is supposed to be always taking place in a gas is known as *ionic recombination*.

#### RECOMBINATION OF THE IONS

That the ions recombine with each other when left to themselves is no mere hypothesis. If a gas which has been ionized is removed from the source of ionization it is found that it soon loses its power to conduct electricity, and if the conductivity of the gas as measured by the saturation current through it be observed at definite intervals of time after removing it from the agent of ionization, the law which governs this decay of conductivity may be studied. This has been done by Rutherford, Townsend, McClung, Langevin and others,

using different experimental methods and different ionizing agents, and the results of their work show that the number of ions present in a gas at a time  $t$  after removing it from the ionizing agent is given by the equation

$$\frac{1}{N} - \frac{1}{N_0} = \alpha t$$

where  $N$  is the number of ions in the gas after a time  $t$ ;  $N_0$  the number of ions originally present in the gas; and  $\alpha$  is a constant known as the coefficient of recombination of the gas.

Now it can be shown that if a gas containing an equal number of positive and negative ions is removed from the source of ionization, then, if the gas loses its power of conducting electricity only on account of the recombination of the ions, the rate at which the conductivity of the gas diminishes would be given by the above expression which, as we have seen, has been verified by experiment. Thus this law is a direct consequence of the ionization theory of conductivity, which it therefore confirms.

The value of  $\alpha$  varies slightly for different

gases, its value for air being about  $3400 \times e$  (where  $e$  is the charge carried by each ion). The value of  $a$  does not vary with the pressure of the gas, being constant over very wide ranges of pressure, although at very low pressures the value of this constant appears to decrease as the pressure is reduced.

McClung has shown that the value of  $a$  increases in a somewhat complicated manner as the temperature of the gas is raised.

#### MOBILITY OF THE IONS

The current through a gas is carried by ions which move through the gas in the direction of the electric field, and in this respect the conduction is analogous to that of electricity through an electrolyte. By a well-known method it has been shown by Kohlrausch that in the latter case it is possible to measure the sum of the velocities of the positive and negative ions by measurement of the conductivity of dilute solutions of known concentration. In a manner somewhat similar to this it is possible to measure the sum of the velocities of the ions in a gas.



If  $C$  be the current per unit area which passes through a gas between two parallel plates, and  $u$  and  $v$  are the velocities of the positive and negative ions respectively, then if there are  $n$  positive and  $n$  negative ions per cubic centimetre each with a charge  $q$

$$C = nq (u + v) \dots \dots (1).$$

Hence the current is proportional to the sum of the velocities of the positive and negative ions. Now if the ionizing agent of the gas be suddenly removed and a large potential difference applied between the plates so that all the ions are driven out before they have time to recombine, then each plate receives a charge  $nq$  if the plates are at unit distances apart. If this charge be measured we have, with the use of equation (1), the means of determining  $u + v$ , and this quantity is found to be proportional to the potential difference between the plates.

The sum of the velocities  $u$ , and  $v$ , with which the positive and negative ions respectively move under a potential gradient of one volt per centimetre is defined as the *mobility* of the ions.

In order to find the values of the mobility of the ions separately it is necessary to find some other relation between  $u$  and  $v$ . It was shown by Hittorf that by analyzing the liquid near the electrodes after the passage of a current through an electrolyte, it is possible to obtain the ratio of the velocity of the positive ion to that of the negative ion in a known electric field. Although this method of chemical analysis is in the nature of things inapplicable to the case of gases, it has been found possible to obtain the same ratio by a method employed by Zeleny. For this purpose two oppositely charged perforated plates are immersed in the gas and a current of air passed through them along the lines of electric force. It will be seen that the positive and negative ions move, under the influence of the electric field, with and against the air current respectively. By reversing the field and readjusting the potential difference between the two plates it is possible to arrange that one of the plates should now receive as much negative electricity per second as it previously obtained positive. Considering the unit area of the plate,

the charges received by the plate in the two cases are respectively  $p u - V$  and  $p v - V$  per second, where  $p$  and  $p'$  are the potential gradients in the two experiments, and  $V$  is the velocity of the air current. And since the experiment is so arranged that these two quantities are equal it follows that

$$\frac{u}{v} = \frac{p'}{p}$$

which gives the relation required.

Although several methods have been used to measure the sum of the velocities of the ions and their velocities separately, sufficient has been said to indicate how these quantities can be determined. The value of the mobility of the positive ion in dry air at atmospheric pressure is 1.36 centimetres per second, and for the negative ion 1.87. The values for other gases are not very different except for hydrogen, in which gas the ions move much more quickly. In damp air the negative ions appear to move more slowly than in dry air, but the velocity of the positive ions is almost unaffected by the presence of mois-

ture. This is a matter of some importance, and indicates that the negative ions have the power of condensing moisture round them.

#### THE CHARGE CARRIED BY THE NEGATIVE ION

It was pointed out above that if we have two parallel plates at unit distance apart, then if there are no fresh ions being produced and the ions present are removed from the gas by an electric field before they have time to recombine, the charge received by the positive plate is  $nq$ , where  $n$  is the number of negative ions present and  $q$  is the charge carried by each ion. The charge received by the plate can be measured, and it is therefore clear that if we had the means of determining the number of ions per cubic centimetre of the gas, we could calculate the charge carried by each ion.

Now, in the case of conduction through an electrolytic solution, we know that 96,000 coulombs, or roughly  $10^4$  electromagnetic units, liberate 1 gramme of hydrogen at normal temperature and pressure, and this amount of gas occupies 11.2 litres. Hence 1 electro-

magnetic unit liberates 1.16 c.c. hydrogen at normal temperature and pressure. Now it has been estimated from indirect evidence derived from the kinetic theory of gases that 1 c.c. of a gas contains between  $2 \times 10^{19}$  and  $10^{20}$  molecules. Taking the mean value  $6 \times 10^{19}$ , we find that 1.16 c.c. hydrogen contain  $6.96 \times 10^{19}$  molecules, and therefore  $1.39 \times 10^{20}$  atoms. Now these atoms carry 1 electromagnetic unit of electricity, which is equivalent to  $3 \times 10^{10}$  electrostatic units. Hence each atom of hydrogen carries  $\frac{3}{1.39} \times 10^{-10} = 2.16 \times 10^{-10}$  electrostatic units.

In the case of the conductivity of electricity through gases, it is not possible to make any similar calculation, since the discharged ions cannot be collected and their volume estimated as in the above case. It has, however, been shown by Professor J. J. Thomson that the number of ions per c.c. in a gas can be indirectly estimated in the following manner:—

When a gas is allowed to suddenly expand it is cooled, and if the gas is, previous to the expansion, saturated with moisture, the ex-

pansion may under suitable conditions cause the formation of a cloud. For a long time it was thought that the drops of water in the cloud were formed round dust particles, and that dust was necessary for the formation of such clouds; but the subject was very thoroughly investigated by C. T. R. Wilson, who showed that a cloud could be formed even in perfectly dust-free air provided that the expansion exceeded a certain value and that the air was ionized. The greater the ionization, the denser was the cloud formed, and if the ions were removed by a strong electric field, the density of the cloud was greatly diminished. Wilson further showed that if the final volume of the gas after expansion exceeded the initial volume by more than 25 per cent but less than 31 per cent, only the negative ions acted as condensation nuclei and not the positive. The drops of water so formed may be easily seen in a beam of light, and although their number per cubic centimetre cannot be directly counted, this quantity can be estimated by measuring the time taken for the cloud to fall. Since the individual

drops of water in a cloud are exceedingly small, it is obvious that they will settle only very slowly on account of the resistance to their motion offered by the air through which they are falling; consequently the smaller the drops, the slower will be the rate at which they settle. Thus the rate at which the cloud falls gives an estimate of the size of the water particles forming the cloud, and if, further, some method can be devised for finding the total quantity of water contained in the cloud, it is a matter merely of calculation to determine the number of drops constituting the cloud. Now it has long since been shown by Stokes that if a small sphere of density  $\rho_0$  fall through a viscous fluid of density  $\rho$ , then the terminal velocity of the drop is given by the equation

$$u = \frac{2}{9} \frac{\rho_0 - \rho}{\mu} g \alpha^2$$

where  $\mu$  is the coefficient of viscosity,  $g$  the acceleration due to gravity, and  $\alpha$  the radius of the drop. In the case under consideration of a globule of water falling through air,  $\rho_0 - \rho$  may be taken to equal  $\rho_0$ , since the density of

air is small compared with that of water. If, then,  $u$  is measured, the radius of the drop can be calculated.

Now it is easy to deduce the *total* mass of water precipitated if the expansion of the gas by which the cloud was formed took place adiabatically, that is to say in such a way that no heat is either gained or lost during the process of expansion. Hence, if we know the mass of each drop we can calculate the number of drops present, and on the assumption that every negative ion acts as a condensation nucleus we can calculate the charge carried by a single ion. The details of the calculation are somewhat complicated and need not here be considered, but without entering into details the general method will be understood from the above account.

Proceeding in this way, J. J. Thomson found  $3.4 \times 10^{-10}$  electrostatic units for the charge carried by the negative ion produced by Röntgen rays and ultra-violet light. H. A. Wilson, by a slightly different method, gives the value as  $3.1 \times 10^{-10}$ . Considering the difficulties of the experiments both these values



agree sufficiently nearly with that obtained for the charge carried by the hydrogen ion in electrolysis, and it may therefore be concluded that these two charges are, in fact, equal.

A.N.S.

#### THE RATIO OF THE CHARGE TO THE MASS OF THE CATHODE RAYS

When a current of electricity is passed through a rarefied gas at a pressure of less than 1 millimetre of mercury a complicated series of phenomena can be seen as the discharge passes. With the various features of the discharge and their significance we are not here concerned, but there are certain phenomena to which we shall have occasion to refer later and which we will consider briefly.

If a current of electricity is passed between two electrodes through the gas contained in a glass vessel which can be evacuated by means of a pump, the character of the discharge gradually changes as the pressure of the gas is reduced. When the vacuum is sufficiently high, a faint stream of light is seen proceeding in straight lines from the negative electrode or cathode which frequently produces a

green phosphorescence on the walls of the glass at places at which the stream impinges. On account of the fact that the luminosity appears to emanate from the cathode it is usually spoken of as the *cathode stream* or *radiation*. Now these rays have been the subject of much investigation and discussion, and their nature was for a long time not understood, but they have ultimately been shown to consist of material particles projected from the cathode with very high velocities; moreover, they have been shown to carry with them a negative charge of electricity; for if the rays are caused to pass through a strong electric field, the stream which previously proceeded in a straight line from the cathode is now bent towards the positive electrode into a curve, as would be expected if it consisted of negatively charged particles projected from the cathode. Furthermore they are deflected by a magnetic field applied at right angles to the direction in which they are being propagated, which should also be the case on the view just mentioned as to the nature of the rays. For it has been shown by Rowland

that a charged particle in motion has the same magnetic effect as a current of electricity, and hence we should expect that this stream of particles, if charged, should be deflected by a magnetic field acting at right angles to the direction of motion of the particles and the magnitude of the deflection to be expected for a given magnetic field can easily be calculated. For if we have a field of strength  $H$  at right angles to the direction of a current  $C$  flowing in a straight wire, then, as is well known, the force per unit length exerted on the wire is  $HC$ .

Now in our case of a moving charged body,  $C = ev$  where  $e$  is the charge carried by the particle and  $v$  is its velocity.

Since the force always acts at right angles to the path of the particle, it does not affect its velocity but only the direction of its motion. Thus if  $r$  be the radius of curvature of the path of the particle, the electro-magnetic force ( $evH$ ) is balanced by the centripetal force  $\frac{mv^2}{r}$  toward the centre of curvature of the path of the particle. We thus have

$$\frac{mv^2}{r} = ev H$$

$$\text{whence } \frac{e}{m} = \frac{v}{Hr} \dots \dots (1)$$

If we measure  $r$  and  $H$  this gives us a relation between  $\frac{e}{m}$  and  $v$ . In order to find them separately it is necessary to find another relation between these two quantities, and this is done by measuring the deflection of the rays by an electric field. Consider the rays passing between two charged parallel plates and let  $F$  be the electric force between the plates, then  $Fe$  is the mechanical force acting on each particle, and hence each particle moves with an acceleration  $\frac{Fe}{m}$  towards the centre of curvature of the path of the particle, and this acceleration we know to be equal to  $\frac{v^2}{r}$ .

$$\text{Hence } \frac{Fe}{m} = \frac{v^2}{r} \dots \dots (2)$$

Combining (1) and (2) we have

$$\frac{v^2}{rF} = \frac{v}{Hr}$$

$$\text{whence } v = \frac{F}{H}$$

$$\text{and } \frac{e}{m} = \frac{F}{H^2 r}$$

The experiments of deflecting the cathode rays by a magnetic and electric field were successfully performed by J. J. Thomson, who obtained the mean values  $\frac{e}{m} = 7.7 \times 10^6$  electromagnetic units and  $v = 2.7 \times 10^9$  centimetres per second, the directions of the magnetic and electric deflections being such as to indicate that the rays carried with them a negative charge.

Now we have learnt that the charge carried by the negative ion produced by Röntgen rays and ultra-violet light is the same within the limit of experimental error as that carried by the hydrogen ion in electrolysis. Hence, assuming the same charge to be carried by the negative particle in the cathode rays, the mass of the particles must be about  $\frac{1}{1000}$  of the mass of the hydrogen ions in electrolysis for which  $\frac{e}{m} = 10^{-4}$  electromagnetic units, for

the value of  $\frac{e}{m}$  found for these rays has been seen to be  $7.7 \times 10^6$  electromagnetic units.

It will be noticed that the important conclusion that the cathode rays consist of a stream of particles whose mass is only an exceedingly small fraction of that of the hydrogen atom, is based on the assumption that the charge carried by these particles is the same as that carried by the hydrogen ion in electrolysis. Now, although it has not been directly demonstrated, there is much indirect evidence to show that this is the case; for there seems to be little doubt that the carriers in the cathode stream convey with them a charge which is the same as that on a negative ion in an ionized gas; and this we have already seen to be equal to the charge carried by the hydrogen ion in electrolysis.

The demonstration of the existence of these minute negatively-charged particles, or, as they are often called, *corpuscles*, which are so much smaller than the lightest known chemical atom, is in itself of the greatest importance;

but we shall see later that this discovery throws light on the phenomena produced by radioactive bodies which give out a radiation similar to the cathode rays.

#### THE RÖNTGEN RAYS

We have seen that when the cathode rays strike the walls of the vessel in which they are being generated they produce phosphorescence, but under ordinary circumstances the rays do not reach the space outside the discharge tube, since they are stopped by the glass of which the tube is made. Now it has been shown by Lenard that if the portion of the discharge tube upon which the cathode rays fall is made sufficiently thin, the rays are able to penetrate the walls and can be detected outside. Thus when a piece of very thin aluminium was cemented over a hole in the discharge tube and the cathode rays allowed to fall upon it, radiations passed through the aluminium, which exhibited all the properties of the cathode rays. The radiation which thus penetrates the aluminium is, however, now of a more compli-

cated nature than are the cathode rays inside; for whereas the latter can be deflected by a magnetic field, it was shown by Professor Röntgen, of Munich, in 1895 that there is outside the tube, in addition to the cathode rays, a type of radiation which cannot be deflected by a magnetic field. These rays are known as Röntgen rays or X-rays.

This was a most important discovery, and it has been since shown that such rays are always produced when the cathode rays are stopped by a solid obstacle. Moreover, these rays can penetrate considerable thicknesses of solid bodies without being absorbed, so that they may often be detected outside an ordinary discharge tube in which the vacuum is sufficiently high, proceeding from the region of the glass upon which the cathode rays strike, for they possess the property of affecting a photographic plate interposed in their path just as ordinary light does. When it is desired to obtain a concentrated beam of these rays, it is usual not to depend on their being produced by impinging on the walls of the discharge tube, but to construct



special tubes in which the cathode rays are caused to fall upon a metallic plate—preferably of platinum—situated opposite the cathode. In these circumstances a beam of Röntgen rays is emitted from the plate, and after traversing the walls of the discharge tube, pass out into the exterior. A tube suit-

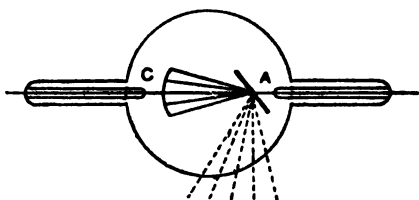


FIG. 2.—Röntgen Ray bulb.

able for the production of the rays is shown in Fig. 2.

The cathode C is made curved so that the cathode rays converge upon the anode  $\alpha$ , which is inclined at an angle of  $45^\circ$ . Consequently from the anode a stream of Röntgen rays is emitted which pass out of the tube as shown by the dotted lines.

Besides their photographic action the rays have the power of exciting fluorescence in

certain substances, notably in a certain class of compounds known as platinocyanides. If, for instance, a screen is coated with a layer of barium platinocyanide and interposed in the path of the rays, the screen becomes brilliantly fluorescent and may be used not only for detecting the rays but also for studying their relative transparency to different substances. For if different obstacles are interposed between the source of the radiation and the screen, those portions of the latter which are protected from the direct action of the rays will appear dark, whereas the rest of the screen will be fluorescent, and according as the obstacle interposed is more or less opaque to this particular kind of radiation the shadow cast on the screen will be darker or lighter. In this way, for example, the shadow of the bones of the hand may be projected upon such a screen. For the rays can pass tolerably freely through the tissues of the hand but cannot penetrate the bones, which therefore cast a shadow of themselves upon the fluorescent screen behind them. If the fluorescent screen is replaced by a photo-

graphic plate, then a shadow photograph or radiograph of the bones will be obtained on developing the plate. If the degree of transparency of the rays to different materials is studied, it is found that, as a general rule, substances of high density are for the same rays more opaque than those whose density is less; but the transparency also depends upon the particular rays used. If the vacuum in the tube from which the rays originate is very high it is found that the Röntgen rays emitted are far more penetrating than when the vacuum is less perfect, so that the results obtained with different tubes cannot be compared.

It appears, therefore, that the Röntgen rays possess many of the properties of light of so short a wave-length as not to be visible to the eye, or as it is usually called *ultra-violet light*; but there is one remarkable difference. It is well known that when ordinary light passes from one medium to another the direction of propagation of the waves of light is changed and the light is said to be refracted. Thus if light passes through a prism of glass

the light which emerges is found no longer to be travelling in the same direction as before it entered the prism. If a similar experiment is made with the Röntgen rays using an aluminium prism which is fairly transparent to the rays, the radiation passes through without being deviated from its original path, so that the rays have not suffered refraction in passing through the prism. This fact and the other properties of the rays have been explained by a theory due to Stokes, according to which the rays consist not of a regular undulatory disturbance propagated through the ether, as is the case with light, but of an irregular series of pulses set up whenever one of the cathode ray corpuscles is stopped. This view is consistent with all the known phenomena connected with the rays, but the equally probable view is held by some that the Röntgen rays consist merely of light waves of exceedingly short wave-length, so that the precise nature of the rays is still uncertain. We shall see later that rays analogous to the Röntgen rays are emitted by many radioactive bodies.

## CANAL RAYS

There is yet another type of rays which can be observed during the discharge of electricity through a rarefied gas and which have their analogue in the rays given out by radioactive substances.

It was shown by Goldstein that if the cathode of a discharge tube in which cathode rays are being produced is perforated by a number of holes, streams of light appear to pass out in straight lines behind the cathode through the holes. These streams were called by Goldstein "Kanalstrahlen," or canal rays. The properties of these rays have been carefully studied, and it was shown by Wien that they consist of positively charged particles moving with high velocities for which the value of  $\frac{e}{m}$  was of the same order as for the hydrogen ion in electrolysis. The rays therefore consist of projected particles charged with electricity of an opposite sign to that carried by the cathode rays, whose mass is of the same order as ordinary atoms of matter, assuming that each particle carries a charge equal to that carried by the hydrogen ion in electrolysis.

## CHAPTER II

### ON THE METHODS OF MEASURING CURRENTS THROUGH GASES

#### THE QUADRANT ELECTROMETER

ELECTRIC currents through gases are usually exceedingly small, and except in cases of very intense ionization it is inconvenient, if not impossible, to measure them by means of a galvanometer by any of the ordinary methods adopted for the measurement of currents flowing in metallic conductors. For this reason special methods have to be employed, and one of the most convenient instruments used for the purpose is the quadrant electrometer. The instrument, a simple form of which is shown in Fig. 3, was invented by Lord Kelvin, and consists essentially of a light piece of metal cut into the shape shown in Fig. 4 and called the needle, suspended in four hollow quadrant-



FIG. 3.—Old form of quadrant electrometer.





shaped metal pieces (Fig. 3), which are supported on insulating pillars. In many of the older types of electrometers these pillars are of glass coated with shellac, but this insulation is very imperfect, and in the more modern instruments is replaced by supports of ebonite or amber. The

needle is suspended by a quartz fibre, or a bifilar silk suspension, and is adjusted so as to be symmetrically placed with respect to the quadrants when they are metallically connected together and therefore at the same

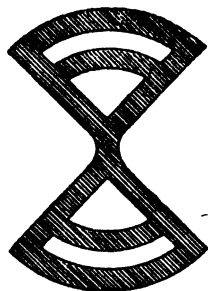


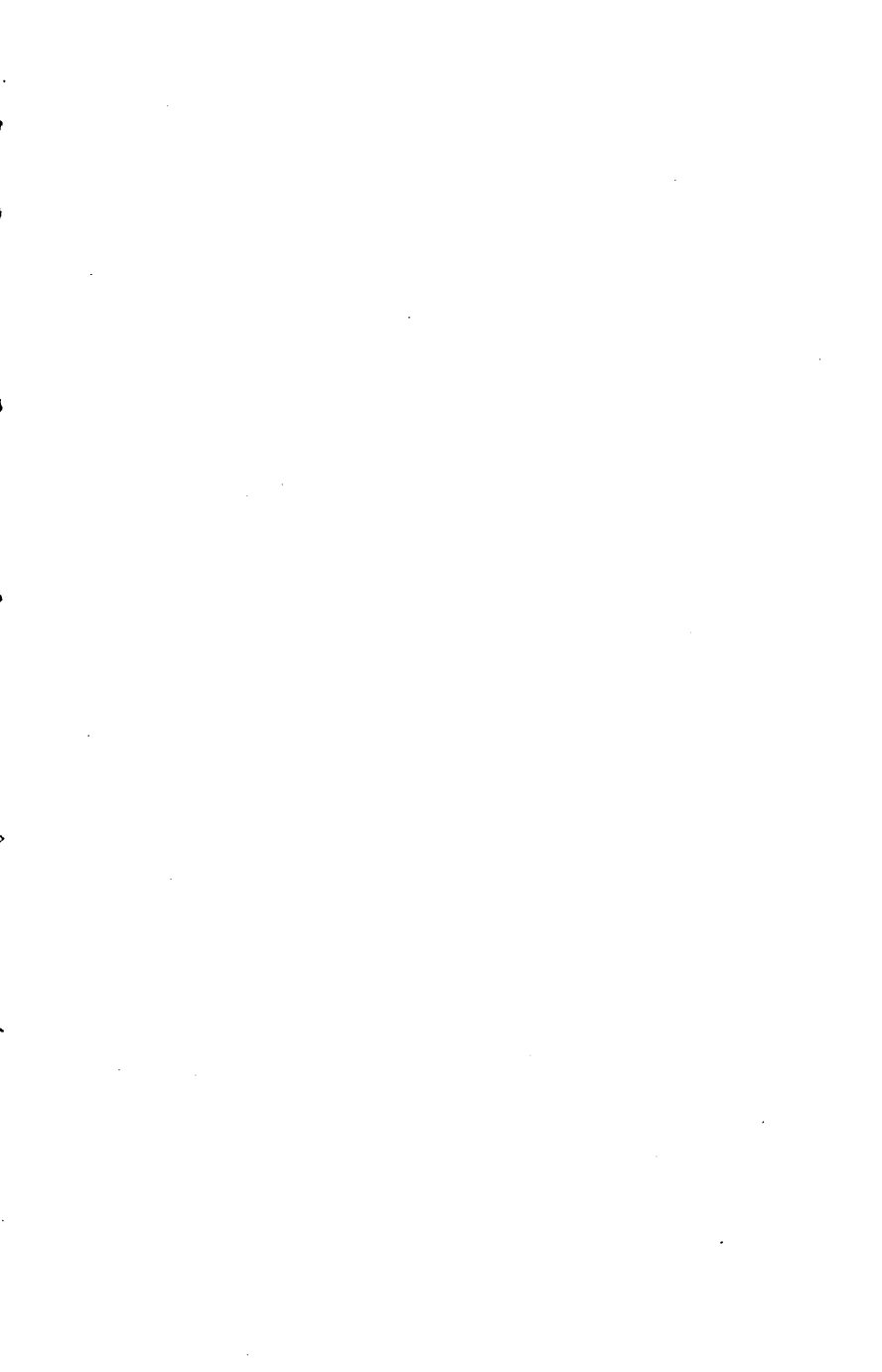
FIG. 4.—Quadrant electrometer needle.

potential. The position of the needle is read by observing the position of a spot of light reflected by a small mirror attached to the needle upon a scale placed at a suitable distance from it. From the needle a platinum wire with a light piece of platinum foil attached to it dips into a glass vessel containing sulphuric acid, which is thus in connection with the needle and increases the

electric capacity of the system. At the same time the platinum foil dipping into the acid serves to damp the motion of the needle when set in motion, and prevents it from swinging backwards and forwards before coming to rest. If the needle is sufficiently light, however, the sulphuric acid may be dispensed with.

To use the instrument the needle is charged to a high potential, and the opposite quadrants are connected by wires. When these two opposite pairs of quadrants are brought to different potentials a couple is exerted on the needle, which tends to draw it into one or other pair of quadrants. This couple is opposed by the twist on the fibre suspending the needle, which eventually takes up a new position of equilibrium depending on the difference of potential between the quadrants. If the instrument has been carefully adjusted the deflection is proportional to the difference of potential between the quadrants, provided that the needle is displaced only through a small angle.

There are other methods of using the instrument, but the one just given is usually



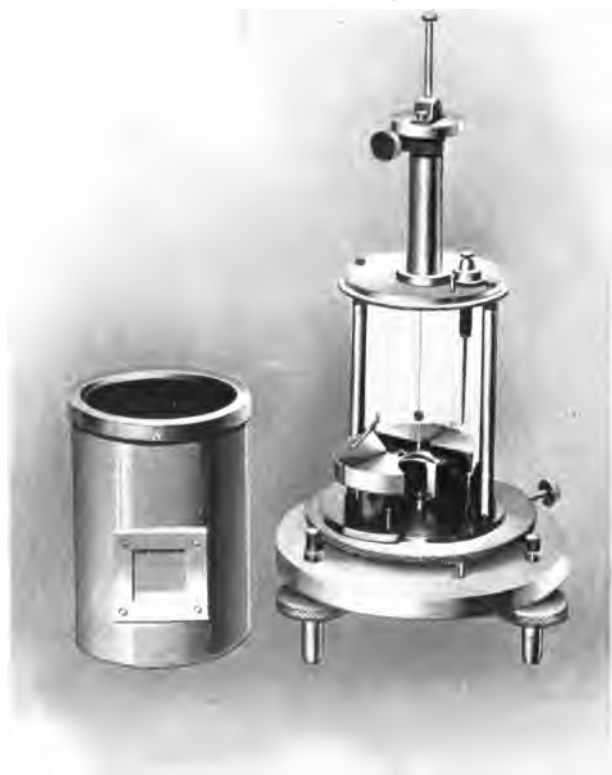


FIG. 5.—Dolezalek type of quadrant electrometer.

the most convenient for measuring currents through gases.

Professor Dolezalek has more recently devised a form of electrometer which is exceedingly simple in construction and which may easily be made very much more sensitive than the older form of the instrument. Such an electrometer is shown in Fig. 5.

The chief modification consists in making the needle of paper coated with a thin film of silver so that it is exceedingly light. In this way it is found possible to do away with the sulphuric acid damping, and to depend on the resistance offered by the air to the motion of the needle to bring it to rest. The needle is suspended by a fine quartz fibre, so that the sensitiveness is greatly increased. It is a matter of no very great difficulty to get a deflection of the needle of 1 metre for 1 volt difference of potential between the quadrants, on a scale placed at a distance of 1 metre from the instrument, which is a sensitiveness exceedingly difficult to obtain with the ordinary pattern of instrument. The quadrants are supported on amber columns which insulate

very perfectly, and the whole instrument is mounted on a revolving table for convenience of adjustment.

USE OF ELECTROMETER TO MEASURE CURRENTS  
IN GASES

The current through a gas is frequently measured between two parallel plates immersed

*QUADRANT ELECTROMETER*

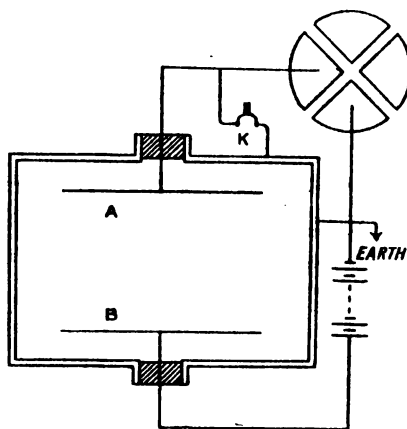


FIG. 6.—Arrangement for measuring current through a gas between two parallel plates by quadrant electrometer.

in the gas (Fig. 6). One of the plates A is connected to one pair of quadrants of the electrometer, the other being connected to

earth. The plate B is connected to a battery of small secondary cells giving sufficient voltage to saturate the current between B and A. One pair of quadrants is kept permanently connected to earth, the other pair being connected to earth by the key K at the beginning of the experiment. On lifting K the current flowing from B to A through the gas causes the quadrants connected with A to charge up and the needle of the electrometer begins to move. The rate of motion of the needle gives a measure of the quantity of electricity passing through the gas in a given time, and therefore of the current. In practice it is best not to observe the needle while in motion, but to allow the current to flow from B to A for a definite time and then to remove the ionizing agent, or if this is impossible to disconnect the quadrants from A. The needle is allowed to come to rest, and the *steady* deflection gives a measure of the quantity of electricity which has passed through the gas in the given time.

In Fig. 7 is shown another form of apparatus which is frequently employed. The plate A

(Fig. 6) is replaced by the rod C (Fig. 7), and the second plate connected to the battery is replaced by a cylindrical vessel coaxial with the rod C. This vessel is connected to one pole of a battery of secondary cells, the other

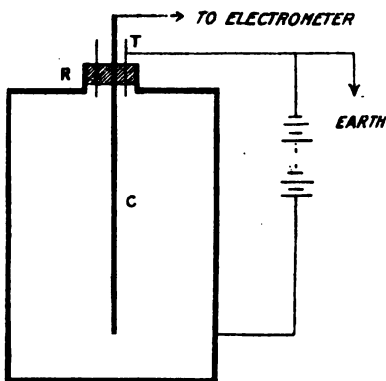


FIG. 7.—Apparatus for measuring current between cylinder and co-axial rod immersed in ionized gas.

pole of which is connected with the earth. The vessel and rod are insulated from each other by the ebonite stopper R, through which passes a metallic tube T connected to earth. This serves to prevent any current flowing across the plug to the electrometer if the insulation of R is faulty.



In making measurements it is necessary to have the electrometer and everything connected with it enclosed in metal connected to earth. It is of the greatest importance to observe this precaution, as otherwise the electrometer receives charges by induction from any electrostatic charge near it or its connections and the measurements become quite unreliable. This precaution necessitates enclosing all wires connected to the electrometer in metallic tubes, and also the keys such as K (Fig. 6) connected to the instrument have to be in metallic cases and raised and lowered from outside by strings attached to them. A very simple and effective way of doing this has been devised by Rutherford. A metallic rod is made to slide in a metallic tube with which it is in contact. The rod dips into a cup of insulating material which contains mercury. When the rod is in the cup the mercury is connected to earth and on raising it by a string the mercury is insulated. The connection from the ionizing vessel to the electrometer is made through this mercury cup.

## ELECTROSCOPE

When a small current through an ionizing chamber of small electrostatic capacity is to be measured it may be found impossible to use a quadrant electrometer to advantage; for if the capacity of the latter is great compared with that of the ionizing vessel, the potential of the plate connected to the electrometer is raised less than would be the case if it were disconnected from the electrometer. For this reason in such circumstances a measuring instrument of small capacity is required, and a gold or aluminium leaf electroscope with small leaves is often a convenient instrument to use. In the ordinary form of electroscope two gold leaves are suspended by a rigid metallic support which is well insulated. On communicating a charge to the leaves they diverge, and the amount of their divergence gives a measure of the potential of the leaves.

In many electroscopes, however, the pair of leaves is replaced by a single leaf and a small rigid plate of metal, and the deflection of the leaf from the plate measured. Such an in-

strument is shown in Fig. 8. A is the rigid metal plate fixed in the insulating stopper I, and L is the moving leaf. The whole is enclosed in a metal box which can be connected to earth, and the position of L is

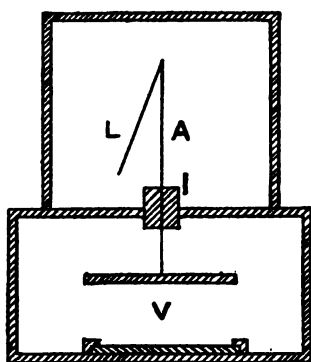


FIG. 8.—Gold-leaf electroscope for measuring currents produced by radioactive substances.

observed through a small window by means of a microscope with a micrometer eye-piece. The rod A is connected to the ionizing chamber V, and the rate at which the leaf L approaches A gives a measure of the current through the former. The instrument may easily be calibrated by charging to different potentials and observing the position of L.

The instrument in this form has the disadvantage that it requires a considerable voltage to deflect the leaf L at all, and the leaf is sensitive to small changes in the value of its potential only when it has itself a considerable potential. C. T. R. Wilson has devised an instrument which is sensitive for very small differences of potential and is often used.

## CHAPTER III

### THE DISCOVERY OF RADIO-ACTIVITY

#### RADIO-ACTIVITY OF URANIUM AND THORIUM

THE discovery that certain bodies spontaneously emitted radiations was the direct outcome of the discovery of the Röntgen rays. We have already mentioned that such bodies are said to be radioactive. As has already been pointed out, if the cathode rays are stopped by the glass walls of the tube in which they are being produced, Röntgen rays proceed from the glass, and at the parts from which they are emitted a brilliant fluorescence of the glass is observed. The view was expressed by M. Henri Poincaré that this fluorescence might in some way be inseparably connected with the emission of the rays, and acting on this suggestion, several physicists directed their attention to investigating whether Röntgen rays were given out by phosphorescent substances such as phos-

phorescent zinc sulphide. Although a photographic record is said to have been obtained by placing this substance on a photographic plate covered by black paper, this result lacks confirmation and cannot be considered as established.

M. H. Becquerel made similar experiments with fluorescent salts of uranium after exposure to light, and obtained photographic impressions on photographic plates wrapped in black paper and exposed to the uranium salts; but after more careful investigation he found that uranium salts acted in this way whether they had been previously exposed to light or kept in the dark, and in fact, that all uranium salts were equally effective and that it was unnecessary to select fluorescent salts at all. It therefore appeared that uranium salts were always capable of affecting a photographic plate and therefore *spontaneously* emitted radiation under all conditions investigated. Uranium itself and all its compounds investigated exhibited this property, the intensity of which did not appear to decrease with time; and it has subsequently

been shown that these rays cannot be reflected, refracted, or polarized, although Becquerel was at first of opinion that this was the case. These rays have been named Becquerel rays after their discoverer and, besides the power of affecting a photographic plate, have the property, not only of penetrating appreciable thicknesses of solid matter opaque to ordinary light, but also of ionizing gases through which they pass; furthermore, they produce fluorescence when they fall upon screens of certain materials, such as zinc sulphide.

Inspired by this important discovery of M. Becquerel, a search was made by several investigators to find out whether any of the other chemical elements exhibited this new property, and it was not long before Schmidt and Madame Curie independently discovered that thorium compounds emitted rays similar in many respects to those given out by uranium; but, as will appear later, there are many important differences between the radiation emitted by these two substances. Quite recently it has been shown, both by Hahn<sup>1</sup> and

<sup>1</sup> O. Hahn, *Proc. Roy. Soc.*, A 76, p. 115.

by Blanc,<sup>1</sup> that the activity of thorium is due, not to the thorium itself, but to a minute quantity of a new substance which can be separated from it by precipitating barium sulphate from a solution containing thorium.

#### RADIOACTIVE MINERALS AND THE DISCOVERY OF RADIUM, POLONIUM, AND ACTINIUM

Two radioactive elements having been discovered it is not unnatural that systematic investigations were made of various minerals known to contain these two elements. Madame Curie first showed that the relative radioactivities of uranium compounds could be satisfactorily measured by the ionization produced in the air between the plates of a condenser by uniform layers of equal thicknesses of the compounds to be compared; if the layers of materials used were uniform, the numbers obtained for the activities of various compounds of uranium were almost independent of the thickness of the layer, the activity of the compound used being in general

<sup>1</sup> G. A. Blanc, *Rendiconti della R. Acad. dei Lincei*, XV, March and April, 1906.



greater the greater the amount of uranium contained by it.

The results obtained with thorium compounds were less consistent, especially when thick layers were used and the activities depended considerably on the thickness of the layers. The cause of these irregularities was subsequently shown by Owens<sup>1</sup> to be due to air currents, and Rutherford<sup>2</sup> has since shown that this action depends on the removal from the thorium of a minute quantity of a radioactive gas which he has called the *emanation*. We shall see later that a similar gas is given off from two other radioactive elements, radium and actinium.

To return to the consideration of the relative activity of various minerals containing uranium. Madame Curie showed that this content of radioactive material varied greatly, and what was of the greatest importance was that certain minerals were more active even than metallic uranium itself. This will be seen from the following numbers which

<sup>1</sup> Owens, *Phil. Mag.*, Oct., 1899.

<sup>2</sup> Rutherford, *Phil. Mag.*, Jan., 1900.

are taken from the results of Madame Curie, where the numbers given represent the current in amperes through the testing vessel used.

Metallic uranium	.	.	.	$2.3 \times 10^{11}$
Pitchblende from Joachimstal	.	.	.	$8.3 \times$
"	"	"	.	7.0
"	"	Cornwall	.	1.6
"	"	Monazite	.	0.5

It thus would seem that the radio-activity of such minerals as pitchblende from Joachimstal cannot be entirely due to uranium, for it would be less radioactive than it is found to be, even if it consisted of pure uranium. This striking discovery was the starting point of the important experiments made by M. and Madame Curie to separate more active substances from these minerals, and the result of which was the discovery and separation of two new radioactive elements, polonium and radium. A third radioactive element, actinium, was subsequently isolated from pitchblende by Debierne.<sup>1</sup>

The method of procedure in separating

<sup>1</sup> Debierne, *Comptes Rendus*, CXXIX, p. 593, 1899 ; CXXX, p. 206, 1900.

these new radioactive elements from pitchblende consisted in subjecting the pitchblende to certain chemical separations and examining the radio-activity of the products obtained by means of an electroscope, those fractions in each separation which showed the greatest radio-activity being reserved for further treatment; in fact, the method was one of ordinary chemical fractionation in which the usual chemical tests were replaced by measurements with an electroscope.

The three new radioactive bodies, polonium, radium, and actinium, are precipitated respectively from uranium ores with bismuth, barium, and certain rare earths, notably thorium. These facts formed the starting point for the separation of these bodies, but as the radioactive elements closely resemble chemically the non-active elements with which they are carried down, their further purification is of considerable difficulty, particularly as the quantities of the substances to be dealt with are exceedingly minute. The method of procedure adopted by Madame Curie was the following.

In the separation of uranium from pitchblende the first process consists in fusing the powdered mineral with sodium carbonate and washing with hot water to which dilute sulphuric acid has been added. The portion which dissolves contains the uranium, and the residue, which is several times more active than metallic uranium, contains the radioactive bodies which are to be separated. The residue consists chiefly of sulphates of lead and calcium mixed with silica, alumina and oxide of iron. Besides these there are present a large number of elements in greater or smaller quantities, and in particular bismuth and barium together with the chemically allied radioactive substances polonium and radium. Actinium is also present in this residue. The radium is present as sulphate, which is even less soluble than the corresponding salt of barium. To redissolve the residue it is necessary to free it from sulphuric acid, which is done by boiling with strong caustic soda. By this means a large part of the calcium, aluminium, and lead sulphates is decomposed, and at the same time alumina, silica, and lead

pass into solution. After washing the residue with water hydrochloric acid is added. The radium remains undissolved, but most of the residue is dissolved by this reagent, and in the solution the new elements polonium and actinium are found. The former can be precipitated by sulphuretted hydrogen and the latter by ammonia. The residue containing the radium is treated with concentrated sodium carbonate, whereby the barium and radium pass into solution as carbonates, which after being carefully washed are converted into chlorides by means of hydrochloric acid. The radium is now reprecipitated by means of sulphuric acid, the solution containing actinium and polonium. The precipitate consists of a mixture of barium, calcium, lead, and iron, with which is mixed the small quantity of radium.

#### RADIUM

To purify the radium, the sulphates are redissolved by boiling with sodium carbonate and again converted into chlorides. A small quantity of polonium can now again be separated by

sulphuretted hydrogen. The solution is treated successively with chlorine and ammonia and the precipitates found to contain some actinium. The filtrate is again treated with sodium carbonate and hydrochloric acid to convert the alkaline earths into the corresponding chlorides. The solution is evaporated to dryness and the residue washed with strong hydrochloric acid. The calcium chloride is dissolved and a mixture of barium and radium chlorides is left behind. To obtain 60 kilogrammes of this mixture it was necessary to treat about 1 ton of mineral. The radio-activity of this residue was about sixty times greater than that of uncombined uranium.

The further isolation consisted in a tedious process of fractional crystallization, and the separation of the radium from the barium depended on the fact that radium chloride is even less soluble in water than barium chloride. The separation may also be effected with advantage, as has been shown by Giesel, by fractionating the bromides instead of the chlorides. As the fractionation goes on, the crystals become more and more coloured if

left to stand, assuming successively as the purification proceeds a yellow, orange, and sometimes a pink colour. When the purification is pushed still further the coloration becomes less marked. This effect is therefore probably due to some action of the radium rays on the barium chloride crystals with which it separates.

By the laborious process sketched above, it was possible eventually to prepare specimens of radium bromide of considerable purity; but the interesting experiment of isolating radium itself remained still to be tried. Although it is possible to study the properties of radium when in a state of combination as a salt, it would obviously be of interest to ascertain whether the peculiarities of this remarkable element were in any way changed by obtaining it uncombined, and also to study its chemical and physical properties.

Now it might be expected a priori that it would be no easy task to separate radium in a metallic state; for in the first place only very minute quantities of its salts are available and the difficulty of the chemical manipulations

to be undertaken are therefore greatly increased. In the second place radium belongs to the group of elements of which calcium, strontium, and barium are members, and is of greater atomic weight than any of these elements. Now it is found that the difficulties of isolating the elements of this group grow as the atomic weight increases. Thus metallic strontium is more troublesome to prepare than calcium, and barium is in turn more difficult to isolate than either. It might therefore be expected that the isolation of radium would present difficulties even greater than those attendant on the preparation of barium even if its salts could be obtained in equally large quantities.

Although the problem has not yet been solved, a step towards the solution has been made. It has been shown by Coehn<sup>1</sup> that if a solution of radium salt is electrolysed under suitable conditions with a cathode of mercury, it is possible to obtain an amalgam with the mercury at the cathode where the metallic radium is separated. When in this state the

<sup>1</sup> Coehn, *Ber. der Deutsch. Chem. Gesel.*, XXXVII, 811. 1904.



radium appeared to exhibit the same radioactive properties as when in the form of a salt.

#### POLONIUM

For the isolation of polonium Madame Curie adopted the following plan.

As we have seen, the polonium is contained in the precipitates with sulphuretted hydrogen in the hydrochloric acid solution obtained during the separation of radium. These precipitates contain bismuth, copper, and lead, and traces of antimony and arsenic. The sulphides are re-dissolved and again precipitated with sulphuretted hydrogen in a strongly acid solution when most of the polonium comes down with the bismuth. The precipitate is then washed to remove any traces of arsenic and antimony, and after washing with water and ammonium nitrate is dissolved as far as possible by the addition of weak nitric acid. The bismuth with polonium is separated in the usual way from copper and lead by precipitating the basic nitrates. The rest of the process consists of a fractional crystallization of the basic nitrates. Polonium has, however,

never been obtained free from bismuth, and it is doubtful whether spectroscopic analysis has revealed any but the lines of the latter metal. The separation is rendered the more difficult by the fact that polonium slowly loses its activity with time, having only half its original activity at the end of about 140 days. This is the first instance that has come to our notice of the loss of activity with time, and we shall have to return to this important property of radioactive substances later.

Marckwald has prepared a substance which is similar in behaviour to the polonium discovered by Madame Curie. The method of preparation consisted in immersing a rod of bismuth in a hydrochloric acid solution of bismuth obtained from pitchblende. In these circumstances the rod becomes coated with a radioactive deposit, the solution at the same time losing its activity. Marckwald considered that the new substance deposited on the bismuth was a body chemically allied to tellurium and called it *radio-tellurium*. This substance has been the object of much dis-

cussion, but there can be little doubt now that it is identical with polonium.

#### ACTINIUM

We saw that polonium was separated out from the other radioactive constituents of pitchblende by precipitation with sulphuretted hydrogen in acid solution. Debierne<sup>1</sup> investigated the portion not so precipitated, and found it to contain a new substance, which has been called by him *actinium*. The material at this stage in the separation contained no uranium, but a certain quantity of polonium and radium still remained mixed with the actinium. The problem was how to separate these two substances, and this was ultimately done by precipitation with ammonia. The precipitates so formed consisted principally of oxide of iron and aluminium, but contained traces of many metals, such as zinc, manganese, and chromium, as well as several rare earths. This crude product, however, contained no polonium or radium. In a later

<sup>1</sup> Debierne, *Comptes Rendus*, CXXIX, p. 593.

publication<sup>1</sup> Debierne gives several methods for separating the actinium in a pure state, and shows that the body separated has properties differing from those exhibited by any of the previously known radioactive elements.

#### ATOMIC WEIGHT OF RADIUM

The determination of the atomic weight of radium has been the subject of a long and careful series of experiments by Madame Curie. The method used was similar to the well-known method used in the case of barium and consisted in weighing the silver chloride obtained from a known weight of the anhydrous radium chloride. The atomic weight was determined from the chloride of radium obtained at various steps in the separation from barium. In the early stages until the activity was only 600 times the activity of uranium, no difference could be detected from the values obtained with pure barium chloride. When, however, the activity had reached 3500 times the activity of uranium, a decided difference began to appear, the mean atomic weight being 140,

<sup>1</sup> Debierne, *Comptes Rendus*, CXXX, p. 906.

which exceeds by an appreciable amount that of barium, which is 137. It therefore appeared that the atomic weight of radium was greater than that of barium.

With the purest specimen of radium chloride obtained, which appeared from spectroscopic examination not to contain more than a trace of barium, the value 225 for the atomic weight was obtained. This number Madame Curie considered to be very near the true value for the atomic weight of radium. A more recent determination gave the value of the atomic weight of radium as 226.5.

#### SPECTRUM OF RADIUM

As has been mentioned, it has been found impossible to detect with certainty a spectrum of polonium, its mixture with bismuth exhibiting only the lines of the latter. With radium the case is different; even when mixed with considerable quantities of barium, lines appear which do not belong to the latter element. With some very impure samples of radium obtained in the first stages of the separation of radium from pitchblende Demar-

cay was able to detect some of the most important spectral lines of radium. His measurements did not, however, extend beyond the ultra-violet and blue end of the spectrum with the exception of one fairly prominent line in the green of wave-length  $\cdot 0005665$  millimetres. The spectrum of radium has since been examined by Berndt, Miethe, Exner, Haschel, and by Runge and Precht.<sup>1</sup> The latter have very carefully mapped the spark spectrum of radium by means of two Rowland concave gratings, the one with 110,000 lines to the inch and radius of curvature 6·5 metres, and the other of 1 metre radius of curvature and 45,000 lines.

The spectrum of radium obtained by Runge and Precht show certain important analogies to those of magnesium, calcium, strontium, and barium, to which the former element bears striking resemblance in chemical behaviour. It had previously been shown by Runge and Paschen<sup>2</sup> that the strongest lines of these elements may be grouped into three pairs

<sup>1</sup> Runge and Precht, *Annalen der Physik*, XIV, 1904, 418–22.  
Paschen, *Ber. den Berl. Akademie*, June 26, 1902.

called by them respectively the line pair of the primary, first secondary and second secondary series. If the position in the spectrum of any pair of lines in either of the series be measured, it is found that the difference in the frequency of vibration (which is proportional to the reciprocal of the wavelength) for the two lines is a constant quantity which we will denote by  $d$ . This distance is sometimes referred to as the line distance. In the first secondary series this is not accurately true, but there always occurs near to one of the lines a second fainter one which has been called a *satellite*, and if this line is taken instead of the stronger line near which it occurs the above-mentioned law may be applied even in this case. The same grouping has been found to be possible for the lines of the spark spectrum of radium.

Now it is well known that the four elements magnesium, calcium, strontium, and barium occur in the same columns in the *periodic* classification of the elements. Runge and Paschen have shown that in passing down this column the value of  $d$ , defined above,

increases in a regular manner as the atomic weight  $A$  of the element considered increases, and the law which they have formulated may be expressed thus:—In each group of chemically related elements the atomic weight varies as some power  $n$  of the distance apart of the two lines of a pair.

$$\text{Thus} \quad A = kd^n$$

where  $k$  is a constant

$$\text{and therefore } \log_e A = \log_e k + n \log_e d.$$

Hence, if the logarithm of the atomic weight be plotted against the logarithm of the distance between the two members of the pair of lines considered, we should get a straight line. This is found to be very nearly the case, and if the law holds for radium we can deduce the value of its atomic weight by extrapolation.

The relation is shown in the table given, and in Fig. 9  $\log A$  is plotted against  $\log d$ .

	Atomic weight ( $A$ )		$\log A$		$d = \frac{10^8}{\lambda}$		$\log d$
Mg. .	24.34	...	1.386	...	91.7	...	1.96
Ca. .	40.1	...	1.603	...	223	...	2.348
Sr. .	87.6	...	1.942	...	801	...	2.904
Ba. .	137.4	...	2.138	...	1691	...	3.228
Ra. .	(258)	...	(2.411)	...	4858	...	3.686



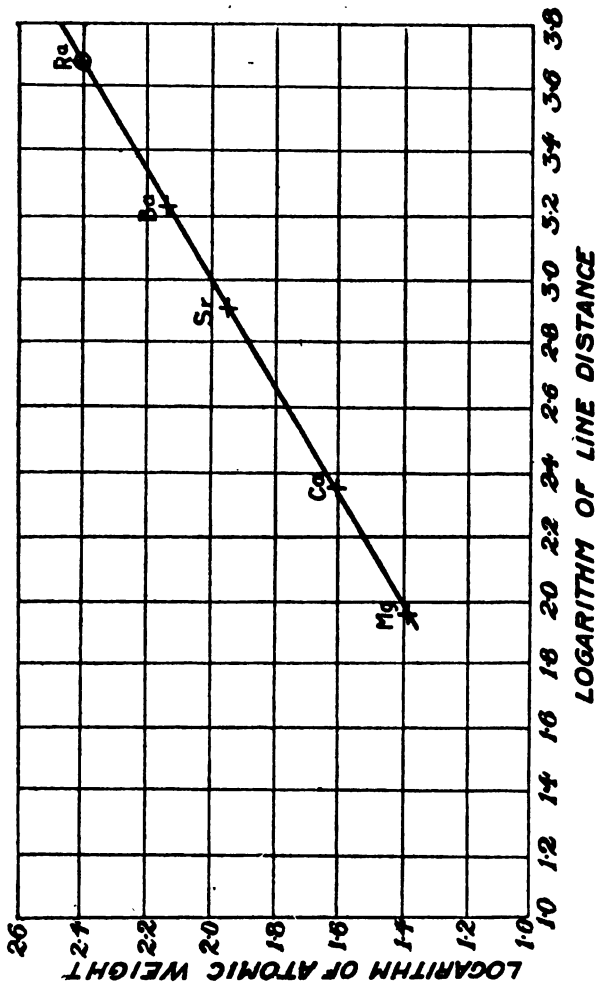


FIG. 9.—Determination of atomic weight of radium by Bunge and Precht's spectroscopic method.



The values of the atomic weight and the logarithm of atomic weight for radium are put in brackets as they have been calculated by extrapolation of the straight line (Fig. 9).

Marshall-Watts<sup>1</sup> has employed a different spectroscopic method of determining the atomic weight of radium, from which he deduces the value 225; Runge and Precht,<sup>2</sup> however, criticize the validity of his method, and it is doubtful whether much weight can be given to this value.

It will thus be noted that there is a discrepancy between the value 226 of the atomic weight of radium obtained by direct chemical analysis and that of 258 obtained by the spectroscopic method of Runge and Precht. This discrepancy still awaits explanation. In the meantime, however, it would seem best to accept the value obtained by direct chemical analysis rather than to depend upon indirect spectroscopic evidence, the interpretation of which is doubtful.

<sup>1</sup> Marshall-Watts, *Phil. Mag.* (6), Vol. V, p. 203 (1903); Vol. VI, p. 64 (1903).

<sup>2</sup> Runge and Precht, *Phil. Mag.* (6), Vol. VI, p. 698, (1903).

## CHAPTER IV

### THE RADIATIONS EMITTED BY RADIOACTIVE BODIES

#### THE $\alpha$ , $\beta$ , AND $\gamma$ RAYS

BODIES which spontaneously give out radiations with definite photographic and electrical actions have been called radioactive, and, in fact, it is only thanks to this property that certain elements such as radium have been detected and isolated at all. Now the radiations emitted by different radioactive elements are not exactly similar, nor are the rays given out by one and the same body in general of one kind; on the contrary, they are often exceedingly complex. That this is the case may be easily shown. If, for instance, some radium be brought into the neighbourhood of a charged gold-leaf electroscope it will be discharged at a definite rate so long as the relative position of the radium and electro-

scope is unaltered. If now an exceedingly thin film of solid matter such as a piece of aluminium leaf is interposed between the radium and the electroscope, the latter is still discharged, but at a somewhat slower rate. It is therefore evident that the rays coming from the radium can penetrate solid objects if sufficiently thin with only a slight diminution of intensity. If now more and more layers of aluminium leaf be interposed until the thickness of the aluminium has reached  $\cdot 005$  millimetres, it is found that the intensity of radiation has been cut down to half its original value. The addition of a further layer of  $\cdot 005$  millimetres of aluminium cuts down the radiation again to half, and so on for successive layers. If, however, this process is continued, it is found that the radiation reaching the electroscope from the radium cannot be completely intercepted, and after a certain thickness of aluminium has been interposed further layers of aluminium have little or no effect until the thickness has been very considerably increased. When the thickness reaches about 1 millimetre the radiation

begins again to be appreciably decreased, and at 5 millimetres is cut down to half. On proceeding still further a similar sequence of phenomena occurs, the radiation ultimately reaching a third constant level. In order to reduce this, it is necessary to interpose even greater thicknesses of aluminium, 8 centimetres being now required to cut down the radiation to half. It thus appears that the radiations are divided into three natural groups: (1) Those which are easily absorbed and which have been called  $\alpha$  rays; (2) those which are less easily absorbed and have been called  $\beta$  rays; and (3) a very penetrating kind of radiation which is known as the  $\gamma$  radiation.

This grouping of the rays into three distinct classes is of great importance, since a further study reveals the fact that the different kinds of rays exhibit important differences besides the dissimilarity in their penetrating power.

Some experiments which are easy to repeat were made by Giesel on the effect of a magnetic field on the radiation from radium. A thin pencil of rays from radium is allowed to pass between the pole pieces of an electro-

magnet at right angles to the lines of magnetic force, and to fall on a phosphorescent screen. A spot of light may be seen on the screen in the dark, and on exciting the magnet the spot of light moves over the screen and the deflection is such as would be produced on a negatively charged particle moving from the radium to the screen. This shows that at least some of the rays from radium consist of negatively charged particles, the magnitude of the deflection being such as to indicate at the same time that they are travelling with exceedingly high velocities. These rays are fairly penetrating, and are the  $\beta$  rays mentioned above.

Although some of the rays are easily deviated by a magnet, yet it was shown by P. Curie that a portion of the rays passed through undeviated, and that it was precisely these rays which are most effective in producing ionization and which are, in fact, the easily absorbed  $\alpha$  rays. A very careful investigation of these rays by Rutherford subsequently revealed the fact that they can be deflected by very *intense* magnetic fields and that they consist of positively charged particles projected with

high velocities. The experiment is, however, difficult to perform, and it should be fully understood that the deflection of the  $\alpha$  rays by a magnetic field is very much less than for the  $\beta$  rays.

The  $\gamma$  rays have so far resisted all attempts to deflect them in a magnetic field, and seem, therefore, to be uncharged, and it seems almost certain that they consist of ether vibrations of some kind and are similar in nature to the Röntgen rays.<sup>1</sup> They have been shown by Marx to travel with the same velocity as light.

The radiations from a radioactive substance may thus be complex, consisting of at most three types of radiation. If the radiation from radium is examined it is found to consist of these three types of radiation; but it does not follow that all radioactive bodies give out all three types. For instance, the rays from polonium consist exclusively of  $\alpha$  rays, there being no penetrating radiation present, so that the  $\alpha$  rays may exist without  $\beta$  or  $\gamma$  rays. In all known cases, however,  $\beta$  and  $\gamma$  rays are found to occur together.

<sup>1</sup> But see note on p. 124.



## THE ALPHA RAYS

MAGNETIC AND ELECTRIC DEFLECTION OF THE  $\alpha$  RAYS

It is only with considerable difficulty that the  $\alpha$  rays can be deflected, and it was some time after the discovery of this easily absorbed radiation that Rutherford<sup>1</sup> at last succeeded

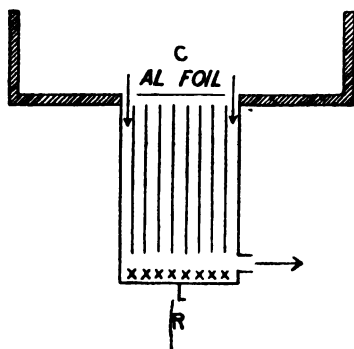


FIG. 10.—Rutherford's arrangement for deflection of Alpha rays.

in deflecting them and demonstrating their true nature. The experiment was performed in the following manner.

The rays from a thin layer of uncovered radium R (Fig. 10) passed up through a number of narrow slits and then through

<sup>1</sup> Rutherford, *Phil. Mag.*, February, 1903.

a thin aluminium leaf into a closed metal chamber C in which the ionization could be measured by means of a delicate gold-leaf electroscope not shown in the diagram. The slits through which the rays passed were situated in the field of a strong electromagnet, the lines of force being at right angles to the plane of the paper. If the rays were deflected by the magnetic field, in their passage through the slits, they would be deflected so as to strike the sides of the slits and be absorbed instead of passing into the chamber C. The rate at which the gold-leaf electroscope lost its charge should thus be reduced if the magnet produced any sensible deflection on the rays. With very intense magnetic fields this was, in fact, found to be the case.

Several precautions were necessary in carrying out the experiment. In the first place it is necessary to prevent the emanation<sup>1</sup> from diffusing into C, for this gas is always given off from radium and is exceedingly radioactive. To prevent this a stream of hydrogen

<sup>1</sup> See p. 51.

was kept passing downwards through the slits as indicated by the arrows in the diagram. If this precaution is omitted the effect of the direct radiation from the radium may easily be completely masked. In the second place it is necessary to make allowance for the  $\beta$  and  $\gamma$  radiation, which passes through the slits and produces ionization independently of the  $\alpha$  radiation when there is no magnetic field. For this purpose the radium was covered with a piece of mica, thick enough to absorb all the  $\alpha$  radiation, but thin enough to allow most of the  $\beta$  and  $\gamma$  rays to pass through unabsorbed. In this way the effect due to these rays alone could be measured and allowed for. Taking these precautions Rutherford was able to deflect nearly the whole of the  $\alpha$  radiation proceeding from the radium.

It now remained to find out in which direction the rays were deflected, and whether the deflection corresponded to a positive or negative charge carried by the  $\alpha$  particles. To do this the slits described in the last experiment were replaced by a set of slits of

the shape shown in Fig. 11, partially closed at the end furthest from the radium. It is clear that a deflection in one direction will now produce a greater effect than an equal deflection in the other, and by reversing the magnetic field it was possible to find out in which direction the rays had been deviated.

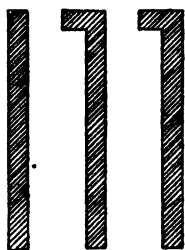


FIG. 11.

As has already been stated the  $\alpha$  rays were found to carry a positive charge. This result has since been confirmed by Becquerel,<sup>1</sup> who used a photographic method for detecting the rays.

It was shown in chapter I. that if the deflection of a charged particle in an electric as well as a magnetic field is known, it is possible to calculate the velocity ( $V$ ) with which the particle is moving, and also the value of  $\frac{e}{m}$  (the ratio of the charge carried by the particle to its mass). It was a matter of even greater difficulty to deflect the rays by an electric field than by a magnetic field, but

<sup>1</sup> Becquerel, *Comptes Rendus*, cxxxvi, p. 199. 1903.

Rutherford was able to obtain this effect and to determine  $V$  and  $\frac{e}{m}$ .

As the result of his early experiment he gives the values :—

$$V = 2.5 \times 10^9 \frac{\text{cm}}{\text{sec.}}$$

$$\frac{e}{m} = 6 \times 10^8$$

It should be mentioned that this value of  $V$  is only the mean value for the rays examined, and there is considerable evidence, to be discussed later, to show that the rays from radium are emitted with velocities varying between definite limits.

Recently Mackenzie,<sup>1</sup> Rutherford,<sup>2</sup> and Rutherford and Hahn<sup>3</sup> have made an extensive examination of the radiation from different radioactive bodies. As a result of their experiments they conclude that, assuming that the particle carries the charge carried by the hydrogen ion in electrolysis, the  $\alpha$

<sup>1</sup> Mackenzie, *Phil. Mag.* (6), X, p. 538, 1905.

<sup>2</sup> Rutherford, *Phil. Mag.*, Oct., 1906.

<sup>3</sup> Rutherford and Hahn, *Phil. Mag.*, Oct., 1906.

particles in every case examined have the same mass and are twice as heavy as the hydrogen atom. This is a result of the greatest importance.

It has long been known that many of the minerals from which the radioactive elements are obtained are precisely those from which the gas helium, discovered by Sir William Ramsay, may be obtained. This gas was shown to have a density of 1.98 compared with hydrogen as unity and to be a mon-atomic gas; its atomic weight is consequently 3.96.

A very important experiment was carried out by Ramsay and Soddy<sup>1</sup> in 1903. The gases evolved by about 30 milligrammes of radium bromide when dissolved in water were collected. These gases were, on analysis, found to consist chiefly of hydrogen and oxygen obtained by the decomposition of the water by the radium. When these gases were removed and the residual gas collected in a very fine capillary tube and examined spectro-

<sup>1</sup> Ramsay and Soddy, *Proc. Roy. Soc.*, LXXII, p. 204, 1903, and p. 346, 1904.

scopically, several lines which could not be identified with those emitted by any known element were observed and these are probably due to the emanation given off by the radium. But if the gas was allowed to stand the helium spectrum appeared, indicating the presence of this gas in the capillary tube. This result has since been confirmed by several workers and may be considered as beyond doubt. The conclusion that helium is being constantly produced from radium follows at once from these remarkable experiments, and the question arises as to what is the origin of this helium.

It was suggested by Rutherford in the Bakerian Lecture to the Royal Society (*Phil. Trans. A*, vol. 204), even before the above-described experiments had been performed, that the  $\alpha$  particles might be charged atoms of helium emitted by the radium with high velocity; and he based his argument on the fact that the mass of the  $\alpha$  particle was not very different from that of helium. But, as has been pointed out above, it has now been definitely shown that, assum-

ing that the  $\alpha$  particle has the charge carried by the hydrogen ion in electrolysis, the mass of the  $\alpha$  particle is half the atomic weight of helium. The  $\alpha$  particle is therefore either not an atom of helium at all or it must have *twice* the charge carried by the hydrogen ion. Rutherford discusses the question and inclines to the latter view; but if the  $\alpha$  particle carries this double charge, it is a remarkable fact and would be quite unique, for it will be remembered that in the case of the ions in a gas produced by various ionizing agents, the charge was always found to be that carried by the hydrogen ion in electrolysis. Hence before this conclusion can be accepted, we must await further experiments on this most important point.

DIRECT MEASUREMENT OF THE CHARGE CARRIED  
BY THE  $\alpha$  RAYS

From the above experiments we may conclude that the  $\alpha$  rays are charged particles whose mass is comparable with the mass of the hydrogen atom, projected with considerable velocity. We should therefore expect that a



radioactive body from which the rays are emitted would, if carefully insulated, become negatively charged as the rays leave it, and that any insulated solid obstacle placed in the path of the rays would become positively charged. If, however, an experiment is performed in which the  $\alpha$  rays are allowed to fall on a solid insulated obstacle just thick enough to absorb all the  $\alpha$  rays but thin enough to allow nearly all the  $\beta$  and  $\gamma$  rays to pass through, the obstacle does not charge up by an amount that can be detected by the most delicate electroscope; nor is the desired effect obtained even if the obstacle and electroscope are placed in the most perfect vacuum obtainable, so as to prevent loss of electricity through the air being rendered conducting by the passage of the rays. This does not agree with the fact that the  $\alpha$  rays can be deflected by a magnetic and electric field, and formed a serious difficulty in the way of accepting the view that the rays really consisted of positively charged particles shot off from radioactive substances, until the discrepancy was explained almost simultaneously by J. J.

Thomson<sup>1</sup> and Rutherford.<sup>2</sup> The experiments of the former were made with radio-tellurium, which gives out only  $\alpha$  rays and which therefore seemed particularly suitable for the investigation. A plate on which a thin layer of this substance had been deposited was fixed 3 centimetres below another plate connected to a sensitive gold-leaf electroscope. The whole apparatus was contained in a vessel which could be exhausted to the best possible vacuum by means of liquid air and charcoal, according to the method discovered by Dewar.<sup>3</sup> When the electroscope was charged *negatively*, the rays falling on the plate produced hardly any effect, the leak being extremely slow; but when charged *positively*, the electroscope was rapidly discharged by the radiation from the radio-tellurium. This is the reverse of what would be expected, and indicated that more negative than positive electricity was reaching the electroscope from the radio-tellurium. The radiation was next made to pass through

<sup>1</sup> J. J. Thomson, *Proc. Camb. Phil. Soc.*, XIII, pt. 1, p. 49, 1905; *Nature*, December 15, 1904.

<sup>2</sup> Rutherford, *Nature*, March 2, 1905.

<sup>3</sup> Dewar, *Proc. Roy. Soc.*, A. LXXIV, p. 122.

a weak magnetic field not strong enough to deflect the  $\alpha$  rays, with a view to separating any rays carrying a negative charge which were capable of being easily deflected. In this way it was, in fact, found possible to stop the negatively charged particles which previously reached the plate connected to the electro-scope, the effect of the  $\alpha$  rays alone could now be studied, and by placing the two plates rather close together, J. J. Thomson showed that the  $\alpha$  rays carried a positive charge. The fact that in the original experiment without the application of the magnetic field the electro-scope lost its charge slightly, even when charged with negative electricity, is explained by the fact that the negative particles moved so slowly as not to be able to reach the negative plate against the repulsion exerted on this by the electric field.

It has been shown by Miss Slater<sup>1</sup> that similar slowly-moving negatively-charged particles are given off by radium emanation. These easily-deflected negatively-charged particles are sometimes known as  $\delta$  rays.

<sup>1</sup> Miss Slater, *Phil. Mag.*, October, 1905.

THE ABSORPTION OF THE  $\alpha$  RAYS

Attention has been drawn to the fact that the  $\alpha$  rays are capable of penetrating appreciable thicknesses of solid bodies without being completely absorbed, and we will now consider the laws governing their absorption by matter. We shall see that the rays are also stopped by gases as well as by solids and liquids, although to a much less extent.

If a layer of radioactive matter is covered by successive layers of aluminium foil, the ionization produced in a testing vessel by the rays penetrating the foil may be measured and compared. This was done by Rutherford and Miss Brooks<sup>2</sup> using radium and polonium as the source of  $\alpha$  rays. The foil was placed immediately over the active preparation, so that the rays traversed no air before reaching the aluminium. For radium the percentage absorption of each successive layer of foil is the same, and the observed ionization falls off in geometrical progression as the absorbing thickness is increased. Thus if  $I_0$  is the in-

<sup>2</sup> *Phil. Mag.*, July, 1902.

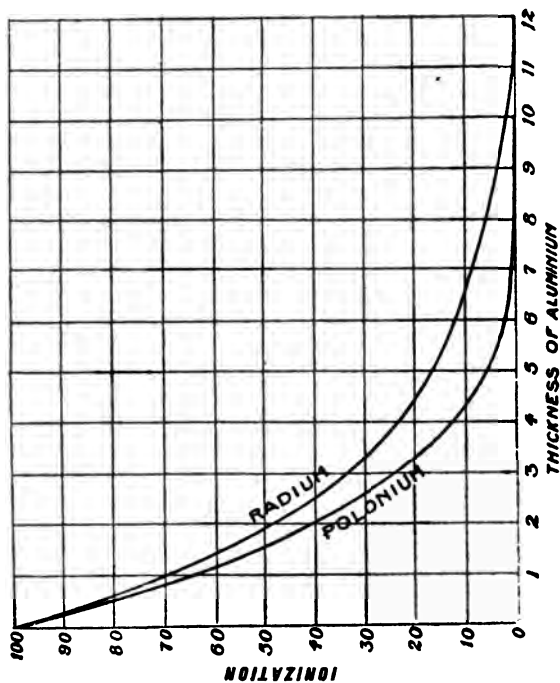


Fig. 12.—Curves showing absorption and penetration of radium and polonium radiations.



tensity of radiation when there is no aluminium foil, then if  $I$  is the intensity when a thickness  $t$  is interposed

$$I = I_0 e^{-\lambda t}$$

where  $\lambda$  is a constant known as the absorption co-efficient which would have a different value if any other material were substituted for aluminium.

This law is true so long as the thickness of the aluminium is not great; but when a certain thickness is reached, the intensity of the radiation penetrating the aluminium falls off very much more rapidly than would be expected from the above law, and quickly becomes zero, showing that if more than a certain thickness of aluminium is interposed *no radiation at all* gets through. The deviation from the exponential law given above is much more marked in the case of polonium, and the curves (Fig. 12) show the relation between the thickness of aluminium penetrated by the rays and the radiation which gets through in the case both of radium and polonium.

As mentioned above, the  $\alpha$  rays are absorbed

by gases as well as by solids and liquids, and it is due to this circumstance that the gas is ionized during the passage of the rays through it. The absorption of the  $\alpha$  rays by air has been demonstrated by Madame Curie<sup>1</sup> in the following way.

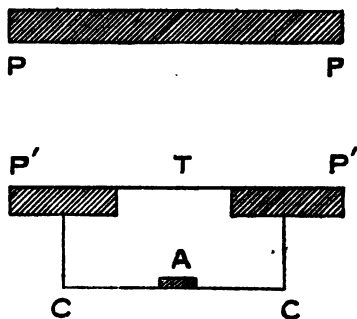


FIG. 13.—Mdme. Curie's experiment on absorption by air of Alpha rays.

Some polonium was placed in a small box C C in the position A (Fig. 13). The rays from the polonium passed up through a piece of aluminium leaf and then ionized the space between the two plates PP, P'P', between which the ionization current could be measured by means of a quadrant electrometer. As the

<sup>1</sup> *Thèse*, p. 49.



distance between A and T was increased, the ionization between the plates fell off, at first slowly and then more rapidly, until when A T was greater than 4 centimetres, no ionization at all was produced between the plates. This, then, showed that the  $\alpha$  rays from the polonium produced no ionization after passing through 4 centimetres of air, and therefore that either the rays were completely stopped by this amount of air or that for some reason they had been deprived of their property of producing ionization. This fact has been the starting point of some most important experiments by Bragg and Kleemann on the "range" or distance which the  $\alpha$  particle can traverse before it ceases to produce ionization.

Before describing these experiments we will first consider the relation between the absorption of the rays and the nature of the material absorbing them.

#### LAW OF ABSORPTION OF THE $\alpha$ RAYS

We have seen that the  $\alpha$  rays are easily absorbed by matter, a few centimetres of gas

being sufficient to completely stop these rays. Now there exists a very simple law between the co-efficient of absorption of the rays and the nature of the gas by which they are absorbed. The absorption of the rays appears to be independent of the physical state or the chemical nature of the gas by which they are absorbed, depending only on *the quantity of matter* which they have traversed.

Thus, for instance, if the pressure of the absorbing gas is raised, the absorption of the rays is correspondingly increased, the absorption being proportional to the pressure of the gas if the temperature remains the same, or, otherwise expressed, *the absorption is proportional to the density of the gas*. If now the absorbing gas is replaced by another gas of different density, not only is the same law found to be true for the new gas when at different pressures, but the relative absorptions of the two gases is found to be in the same ratio as their densities, so that if we know the co-efficient of absorption of any gas, the co-efficient of absorption of any other gas can be calculated from a knowledge of the

relative densities of the gases. It should, however, be mentioned that hydrogen forms a remarkable exception to this rule. Now this law of density is not confined to the case of gases alone. If two substances whose density differs as greatly as those of air and aluminium are compared, the law is still found to hold with a remarkable degree of accuracy. We find that, for example, for the  $\alpha$  rays from uranium, the co-efficient of absorption of aluminium is 1700 times as great for aluminium as for air at normal temperature and pressure; thus taking the density of aluminium as 2200 compared with air at normal temperature and pressure as unity, we see that the co-efficient of absorption is roughly proportional to the density even over this enormous range of density. It should, however, be understood that there are some marked exceptions to this law, which should therefore be applied only with due caution.

#### THE "RANGE" OF THE $\alpha$ PARTICLE

The experiments of Madame Curie have shown that when the  $\alpha$  particle has traversed

a definite distance of air it very suddenly loses its power of ionizing gases, and, as has already been mentioned, Bragg and Kleeman have greatly added to our knowledge of the cause of this effect.

According to the view first expressed by Bragg and Kleeman in a communication to the Australian Association for the Advancement of Science in 1904, and elaborated in subsequent publications,<sup>1</sup> the  $\alpha$  particle was supposed to be absorbed in its passage through a gas on account of the ionization which it produced in its path; and this was considered to be the sole cause of the absorption. To ionize a gas a certain quantity of energy is necessary, which is supposed to be derived from the kinetic energy of the  $\alpha$  particle, which is therefore gradually stopped. In its path the  $\alpha$  particle was supposed to suffer no deviation until its initial velocity had been considerably diminished; thus on this view there should be no scattering of the rays in

<sup>1</sup> Bragg, *Phil. Mag.*, Dec., 1904; Bragg and Kleemann, *Phil. Mag.*, Dec., 1904; Bragg and Kleemann, *Phil. Mag.*, Sept., 1905.

their passage through matter, and they should throw as sharp shadows of opaque objects after as before their passage through matter. This has been shown to be the case by Becquerel using a photographic method.

Theoretical considerations afford this hypothesis much support, and it is possible to deduce the density law, given above, on this view. Moreover, the manner in which the  $\alpha$  particle *suddenly* ceases to produce ionization after its passage through a definite thickness of matter receives a ready explanation. Bragg and Kleeman were able to give a strong proof of the correctness of this view just enunciated by examining the ionization produced in the air between two parallel plates placed very close to each other when placed at different distances from the radiating source. Their early experiments were made with thick layers of radium deposited on a plate; in their later experiments they tried exceedingly thin layers deposited from a strong solution of radium bromide on a small silver dish.

Before giving the results we will consider what should happen as the ionization chamber

is removed from the radiating source. Consider a layer of radium (Fig. 14) so thick that the  $\alpha$  rays coming from layers in the interior suffer appreciable absorption in passing out into the air above. If we assume that each  $\alpha$  particle is projected with the same initial velocity

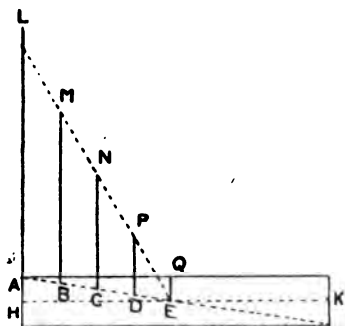


FIG. 14.

from the radium, any particle starting from a point A on the surface of the layer will have a range A L before it ceases to produce ionization. A particle coming from some point B below the surface will have a shorter range B M, and similarly for points C D, at lower depths below the surface. When the  $\alpha$  particle comes from some point E at a depth A H below the surface it will fail to produce any

ionization in the air outside the radium, and all layers below the plane H K will produce no effect. If we further assume that each  $\alpha$  particle produces the same number of ions in each centimetre of gas that is traversed, and

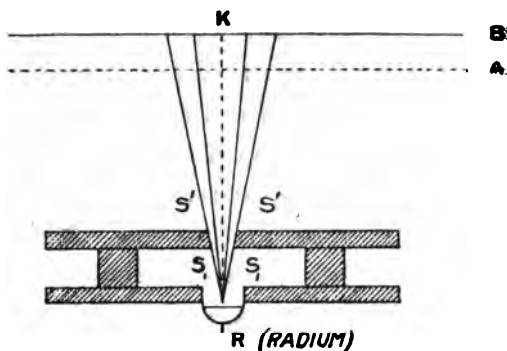


FIG. 15.—Bragg's apparatus for determining range of Alpha particles.

that it loses a definite amount of its energy in so doing, then it follows that the ionization will diminish as a linear function as the distance from the radium increases.

The experiment was carried out by means of the apparatus shown in figure 15. A narrow cone of rays from the radium contained in a small depression passes upwards through the

two slits S S, passes through a wire gauze plate A and falls upon the metal plate B placed close above A. In passing through the space between A and B the air is ionized, and the amount of the ionization may be measured in the usual way by observing the saturation current between A and B. A and B were placed at a distance of about 1 millimetre apart, so that this current could be taken as measuring the ionization at the distance R K from the radium. By moving the ionization chamber A B vertically up and down the ionization at points at various distances from the radium could be measured. If the theory we are considering is correct, then if the ionization is plotted against the distance R K we should get a straight line.

As a matter of fact this is found to be the case if the experiment is performed over limited ranges, the whole curve being made up of *four straight lines* AB, BC, CD, DE, inclined at different angles to each other (Fig. 16). This is what would be expected if radium emitted four sets of  $\alpha$  rays each with definite ranges. There is much direct and indirect



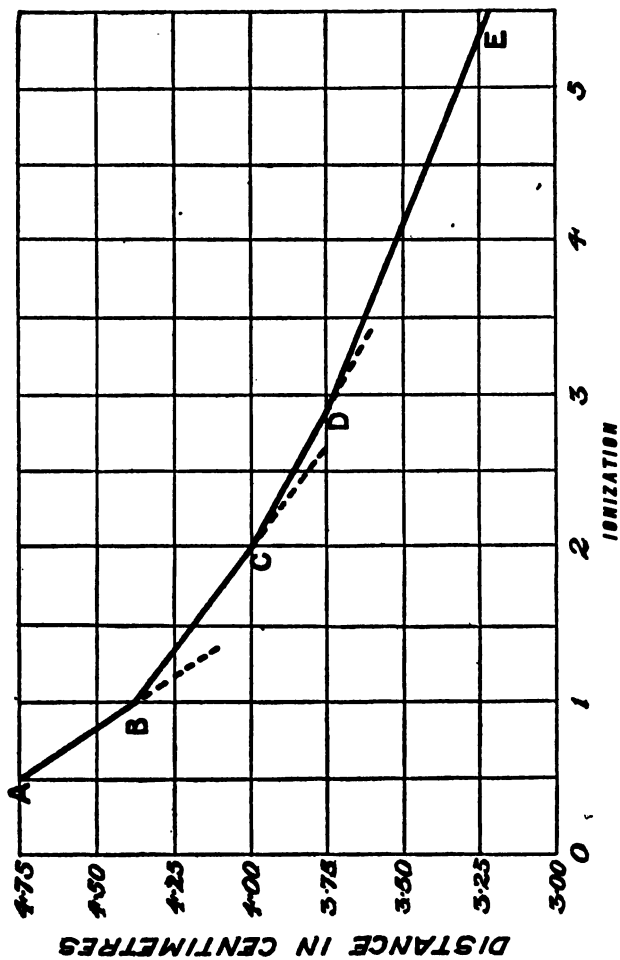


Fig. 16.—Variation of ionization of air by Alpha rays with distance from radium.

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evidence that this is the case. The experiments made with thin layers of radium substantiate this conclusion and are even more striking than the experiments just described.

In the case of a thin layer of radioactive matter, there will be little, if any, absorption

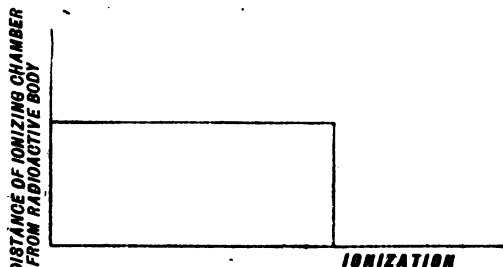


FIG. 17.

of the rays in the radioactive material itself. If, then, there are emitted  $\alpha$  rays of only one definite range, then as the ionization chamber is removed we should expect the ionization to remain constant until a certain distance is reached where the ionization should fall suddenly to nothing. The effect to be expected is shown graphically in Fig. 17.

Now radium can be temporarily brought

into a condition in which it emits  $\alpha$  rays of only one range by heating it to redness for a short time. With a coating of radium bromide which had been so treated and which was spread in a thin layer on a metal plate Bragg and Kleeman obtained the curve shown (Fig. 18). The steep part of the curve C was due to a small ionization produced by  $\beta$  and  $\gamma$  rays, from which it is difficult to *completely* free the radium even by prolonged heating.

A fact that is very noticeable is that in the region A of the curve the ionization produced by the  $\alpha$  particle *increases* as it is removed from the radium from which it is projected. Thus as the velocity of the  $\alpha$  particle is diminished it seems at first to become a more effective ionizer, after which it *suddenly* loses its power of producing ions at all, as is seen from the portion of the curve B.

On allowing the radium to stand for a month it gradually regained the power which it possesses of giving out  $\alpha$  particles with four distinct ranges, and the curve (Fig. 19) shows the behaviour of the preparation after this

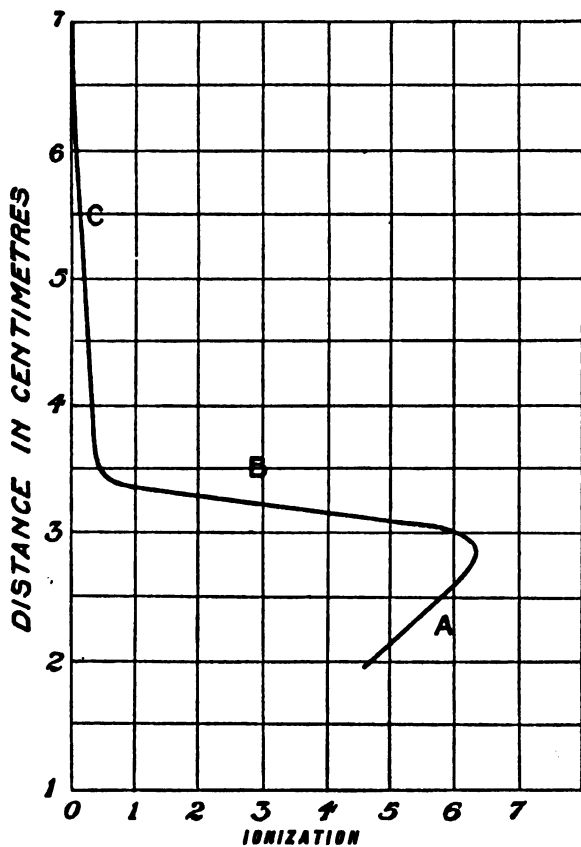
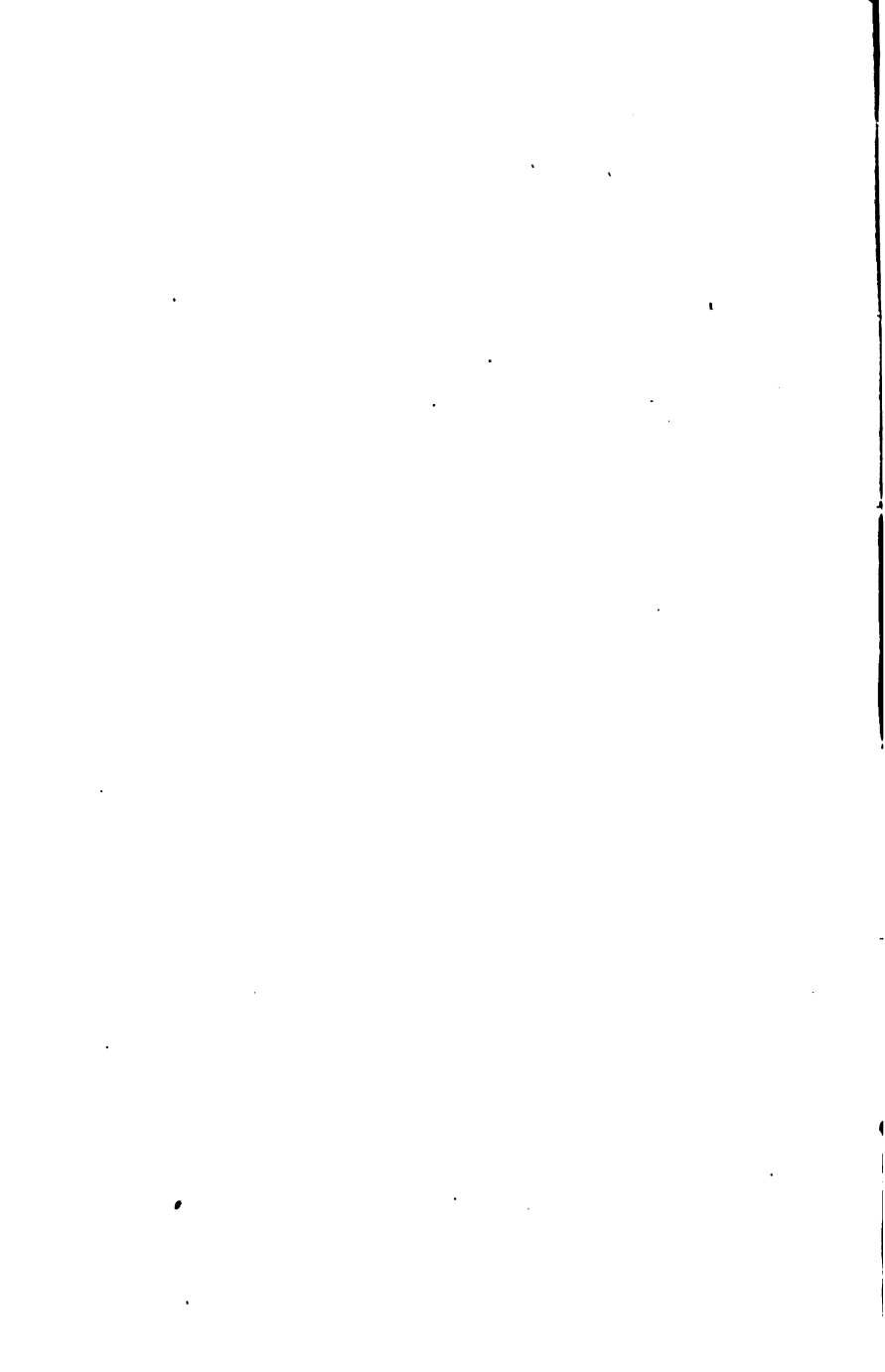


FIG. 18.—Range of Alpha particle from preparation of radium strongly heated just before observation.



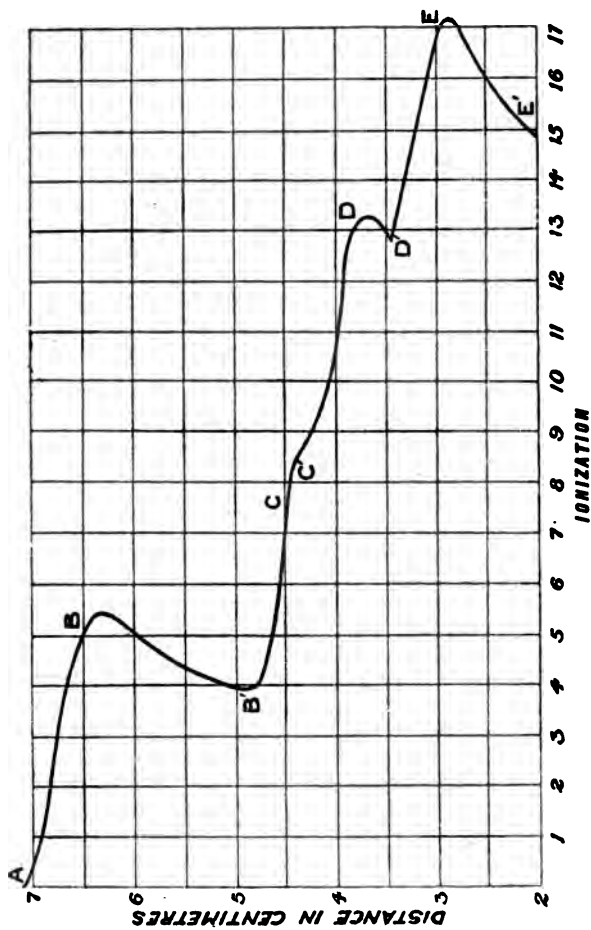


FIG. 19.—Diagram of four groups of Alpha rays of different range emitted by radium.





time. We shall see later that the three sets of rays which thus reappear in the radiation from radium are due to *three definite radioactive products* which are constantly being produced from radium, and it would seem that each of the products gives out rays whose "range" has a definite value, so that the existence of a set of  $\alpha$  rays of a definite range gives evidence of the probable existence of a definite radioactive product.

The  $\alpha$  radiations from uranium, thorium, actinium have been examined by various investigators in a manner similar to that described above, and much useful information obtained as to the nature of the processes taking place in these bodies.

Professor Rutherford<sup>1</sup> has more recently shown that when the velocity of one of the sets of  $\alpha$  rays from radium has been reduced by only 40 per cent of its initial value the rays cease to produce ionization. His experiments consisted in measuring the velocity of the rays in the usual manner after passing through successive layers of aluminium foil. It was

<sup>1</sup> Rutherford, *Phil. Mag.*, July, 1905.

also shown that the phosphorescent and photographic actions of the rays ceased simultaneously with the cessation of the ionization. Now these three properties afford the only test known for detecting the existence of rays; so that we have the important fact that  $\alpha$  rays travelling with more than half the initial velocity of the particular rays examined could be detected by no known means. Hence there may be such  $\alpha$  rays emitted from many bodies with velocities below this critical value, of which we are unconscious. The property of emitting such radiations may therefore be possessed by more substances than the few radioactive bodies with which we are acquainted.

#### THE SPINTHARISCOPE

It is well known that the  $\alpha$  rays when they strike certain substances produce phosphorescence. Thus a screen on which has been spread a film of zinc sulphide becomes luminous when exposed to their action. This phenomenon has been carefully studied by Sir William Crookes, who examined microscopi-

cally the surface of the zinc sulphide when exposed to the action of the  $\alpha$  rays. The surface was seen to be lit up by numerous bright patches which seemed to flash up and disappear in different positions. This effect appears to be due to the impact of the  $\alpha$  particles on the surface. Whenever an  $\alpha$  particle strikes the surface a flash of light is produced at that point, and it is the result of the combined effect of these minute flashes which makes the surface luminous when viewed by the naked eye.

To illustrate this, Crookes has invented an instrument which he calls the spinthariscopes (*σπινθάρης* = a spark). At one end of a tube is placed a piece of metal, on the surface of which is a small quantity of radium bromide. At a distance of one or two millimetres from it is placed a small screen of phosphorescent material, which is viewed from the other end of the tube through a microscope. In the dark the screen may be seen illuminated by innumerable scintillations appearing, now in one, now in another, position of the field of view.

## THE BETA RAYS

It has already been pointed out that the  $\beta$  rays consist of negatively charged particles projected with great velocity, and that they are very much more easily deflected by a magnetic field than the  $\alpha$  radiation.

MAGNETIC AND ELECTRIC DEFLECTION OF  $\beta$  RAYS

The magnetic deviation of the  $\beta$  rays has been studied in detail by H. Becquerel. The rays from some radioactive body contained in a deep hollow cylinder of small internal diameter passed through a slit and were allowed to impinge on a photographic plate placed above it, so that all the rays struck the plate at the same angle. A sharp image of the slit was formed on the plate; but on deflecting the rays by a magnetic field the image of the slit was found no longer to be bounded by sharp edges, but on the contrary to be spread out into a broad band, showing that the rays had been unequally deflected. This effect was traced to the fact that the rays were emitted with different velocities, thus being deviable to a different extent in

a magnetic field. Becquerel was also able to show that the rays which were most deflected and which were consequently projected with the least velocity, were most easily absorbed by matter.

It was further shown by Becquerel that the  $\beta$  rays are capable of being deflected by an electric field as well as a magnetic field, and experiments were made by him in which these deflections were measured in fields of known strength. Now, as has already been shown, we can, from these observations, at once deduce the ratio of the charge carried by the particles to their mass ( $\frac{e}{m}$ ) and also their velocity. Proceeding in this way, Becquerel showed that the velocity with which the  $\beta$  rays travel is about half the velocity of light ( $3 \times 10^{10}$  centimetres per second), but that for some of the rays the velocity greatly exceeded this value, approaching very nearly to the velocity of light. The value of  $\frac{e}{m}$  was about  $10^7$  or very nearly the same as for the cathode rays.

It thus appears that the  $\beta$  rays emitted by radioactive bodies resemble the rays given off from the cathode when a discharge is passed through a gas at low pressure; for these have also been shown to carry a negative charge, to be deflected by both a magnetic and an electric field and to have the same value of  $\frac{e}{m}$  as the  $\beta$  particles. It should, however, be noticed that the velocity of the latter greatly exceeds that of the former, cathode rays travelling with a velocity greater than  $10^{10}$  centimetres per second having as yet never been obtained. The  $\beta$  rays may therefore be regarded as cathode rays travelling with exceedingly high velocity, and thus consisting of a stream of negatively charged particles whose mass is about  $\frac{1}{1000}$  of the mass of the hydrogen atom.

From theoretical considerations, into which it is here impossible to enter, it may be shown that an electrically charged particle moving with a velocity comparable with the velocity of light behaves as if its mass were greater than when moving slowly. Now we have

seen that the  $\beta$  rays are charged particles moving with different velocities approaching the velocity of light. These rays, then, seem to afford a means of testing experimentally the validity of this important theoretical deduction, the correctness of which has been demonstrated by Kaufmann.<sup>1</sup>

The rays from a small quantity of very active radium passed between two brass plates rather more than 1 mm. apart and between which a high potential of about 5000 volts could be applied. Sparking through this gap was prevented by placing the whole apparatus in a good vacuum. The rays next passed through a very narrow tube, and then impinged on a photographic plate covered with enough aluminium leaf to stop the  $\alpha$  rays. The apparatus was so arranged that it could be placed between the pole pieces of an electro-magnet and a magnetic field applied *in the same direction* as the electric field. If the rays were allowed to fall on the photographic plate without either the electric or magnetic field being applied, a small spot

<sup>1</sup> Kaufmann, *Göttingen Nach.*, Nov. 8, 1901.

on the plate was affected at the point at which the beam of rays impinged. On applying the electric and magnetic fields, the  $\beta$  particles suffered deflections in two directions at right angles to each other, and the impression on the plate was transformed into the curved line shown in the diagram (Fig. 20), since the rays

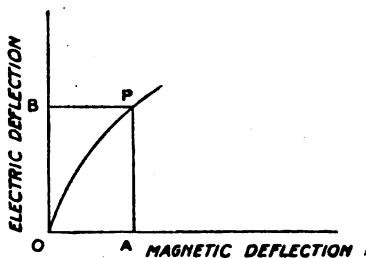


FIG. 20.—Deflection of Beta particles by electric and magnetic fields.

consisted of particles emitted with different velocities, and therefore suffered different deflections in passing through the two fields. By measuring the co-ordinates  $OA$  and  $OB$  for any definite point  $P$  on the curve, it was possible to find the electric and magnetic deflection suffered by the particular ray which struck the plate at  $P$ . As we have already seen, these data give us all that is required



to calculate the velocity of the particle and the value of  $\frac{e}{m}$ . This was done by Kaufmann, who showed that the value of  $\frac{e}{m}$  varied *continuously* as the velocity of the particle increased. The value of  $\frac{e}{m}$  is almost independent of the velocity until the velocity becomes exceedingly near that of light, after which it increases rapidly. It is not till about half the velocity of light is reached that an appreciable change takes place in the value of  $\frac{e}{m}$ . For particles whose velocity was 5 per cent less than the velocity of light the value of  $\frac{e}{m}$  was only one-third of the value obtained for the cathode rays.

It is difficult, according to the most modern views on the atomic nature of electricity, to imagine that the value of the charge carried by the particles can vary *continuously* as the velocity increases; on the other hand, as has been mentioned, there is considerable theo-

retical ground for believing that the apparent mass of an electrified particle should change as its velocity approaches that of light in the manner actually observed. This is the interpretation of the experimental results now generally accepted.

DIRECT DETERMINATION OF THE CHARGE CARRIED  
BY THE  $\beta$  PARTICLES

If the  $\beta$  rays are absorbed by a metal plate upon which they are allowed to fall, they should give up their negative charge to the plate, and this charge should increase continuously until the loss of electricity by the plate due to faulty insulation exactly balances the gain from the  $\beta$  rays. If the plate is connected to a sensitive quadrant electrometer, evidence of this effect should be obtained and the electrometer should become charged with negative electricity. The experiment was tried by Madame Curie, but at first without success. The reasons of failure were twofold. In the first place it is necessary to absorb the  $\alpha$  rays by a thin metal leaf before allowing them to reach the plate connected to the

electrometer, for these rays carry a positive charge and would tend to mask the effect which is sought. In the second place, the rays in their passage through the air before falling on the plate connected to the electrometer, ionize the air, so that the electrometer is no longer well insulated and the charge received by it is dissipated through the air as fast as it is received. To obviate this source of error there are two courses possible; either to remove the air and perform the experiment in a very perfect vacuum, or to imbed the metallic plate in a solid insulator; for though such insulators are rendered conductors of electricity by the  $\beta$  rays, yet they conduct far less than a similar volume of air exposed to the same rays.

This latter plan was adopted by Madame Curie, who was then able to demonstrate by direct experiment that the  $\beta$  particles carried a negative charge with them.

The former method was successfully applied by Strutt. If some radioactive body which emits  $\beta$  rays is suspended by an insulating support in a vacuum, then if the radioactive

substance is confined in an enclosure the walls of which are thin enough to allow the  $\beta$  rays to escape, but thick enough to absorb the  $\alpha$  rays, then the radioactive body should continuously lose a negative charge, or what is the same thing, should charge itself up with positive electricity.

In order to demonstrate this effect Strutt enclosed a small quantity of radium bromide sealed in a glass tube in a very perfect vacuum. The glass tube was supported by a quartz rod and attached to a small gold-leaf electroscope. As the rays escaped from the radium the electroscope gradually charged up. After diverging a certain distance one of the leaves was made to touch a metal plate connected to earth so as to discharge it. The leaves then fell together and recommenced to diverge. The electroscope continued to perform this cycle as long as the vacuum and insulation were maintained satisfactorily. The experiment is not difficult to carry out, and the leaves may easily be made to perform a complete cycle in about 30 seconds. The time of the cycle is independent of all external conditions

and serves as a time measure. For this reason Strutt has called the arrangement a "radium clock."

#### ABSORPTION OF $\beta$ RAYS BY MATTER

The  $\beta$  rays have been seen to be much more penetrating than the  $\alpha$  rays, it being necessary to interpose an obstacle about 100 times as thick as was required for the  $\alpha$  rays to cut down the  $\beta$  radiation by a definite amount. The nature of the  $\beta$  rays, moreover, is very different from that of the  $\alpha$  rays, the charge carried by the particles in the two cases being of opposite signs, and the masses of the carriers very different. In spite of these dissimilarities, the laws of absorption for the two kinds of radiation present many similar features. In the first place, if rays of uniform velocity are used, the law according to which the radiation falls off as the thickness of matter is increased is found to be similar to that for the  $\alpha$  rays, and may be expressed by the formula

$$I = I_0 e^{-\lambda t}$$

where  $I$  is the intensity of radiation after passing through a thickness  $t$  of the absorbing

material.  $I_0$  is the intensity before reaching the absorbing material and  $e$  is the base of the Napierian logarithms;  $\lambda$  is a constant depending on the nature of the matter by which the rays are absorbed.

If the value of  $\lambda$  is measured for different substances it is found to be directly proportional to the density of the substance used, just as was the case for the  $\alpha$  rays.

One or two examples taken from some experiments of Rutherford will suffice to indicate the degree of truth in this law.

Substance.	$\lambda$	Density.	$\frac{\lambda}{\text{Density.}}$
Wood . . .	2.16 ...	.40 ...	5.4
Ebonite . . .	6.5 ...	1.14 ...	5.6
Aluminium . . .	14.0 ...	2.6 ...	5.4
Iron . . .	44 ...	7.8 ...	5.6

From the values given it will be seen that the value of  $\lambda$  divided by the density of the substance is appreciably constant for substances as unlike each other as wood and iron.

There are, however, certain important exceptions to this law, the most notable being lead and tin, for which the values of  $\frac{\lambda}{\text{density}}$

are respectively 10.8 and 13.2. Thus these two metals absorb the  $\beta$  rays more powerfully than the law considered would predict.

The value of the co-efficient  $\lambda$  depends upon the velocity of the rays, being very much greater for rays of small velocity than for those of high velocity.

#### THE GAMMA RAYS

We have yet to consider the third and very penetrating group of rays emitted by radioactive substances and which we have called the  $\gamma$  rays. These rays can penetrate several centimetres of lead without being completely absorbed, and cannot be deflected even by the strongest magnetic or electric field. The  $\alpha$  and  $\beta$  rays frequently appear separately and seem not to be directly related to each other in any very simple manner. The  $\gamma$  rays, on the contrary, seem to be intimately connected with the  $\beta$  rays; these two types of rays always appearing together, the intensity of the  $\gamma$  rays being roughly proportional to that of the  $\beta$  rays. The nature of the  $\gamma$  rays has been the subject of much discussion, and two

hypotheses have been framed to account for their behaviour.

According to the first view, the rays are supposed to consist of corpuscles projected with velocities approaching the velocity of light even more closely than the most penetrating  $\beta$  rays. Now there is reason to suppose that such rays would be very penetrating on account of their great kinetic energy, and exceedingly difficult to deflect in a magnetic field on account of their high velocity. They would thus behave very much as the rays are found to behave. There are, however, several objections to this view.

In the first place we should expect to find rays of all grades intermediate between the  $\beta$  and  $\gamma$  rays becoming more and more penetrating and difficult to deflect in a magnetic field as the velocity increased. As a matter of fact this is not the case, and there is a sharp line of demarcation between the most penetrating  $\beta$  rays, which however can be deflected, and the  $\gamma$  rays, which are entirely incapable of being deflected. In fact, it seems difficult to account for such a *discontinuity* in the proper-



ties of the two sets of radiation if they are essentially similar in nature.

There is perhaps a still more serious difficulty in the way of accepting this theory. If the  $\gamma$  rays convey with them an electric charge, then it should be possible to detect this charge directly just as in the case of the  $\beta$  rays. All attempts have, however, hitherto failed to reveal this effect, although some experiments by Paschen<sup>1</sup> seemed at first to suggest that there was such a charge carried by the rays. But it has since been shown by Eve<sup>2</sup> and others that at points where the  $\gamma$  rays strike a solid obstacle, *secondary* rays of the  $\beta$  type are set up, and it is possible to trace the charges communicated to bodies upon which the  $\gamma$  rays fall to this secondary radiation.

A second hypothesis to account for the properties of the  $\gamma$  rays is that they consist of a very penetrating type of Röntgen rays. We have already seen that the Röntgen rays carry no charge with them, and they cannot

<sup>1</sup> Paschen, *Ann. der Physik*, Nos. 6 and 7, 1904.

<sup>2</sup> Eve, *Phil. Mag.*, Nov., 1904.

be deflected in a magnetic field ; they probably consist of some kind of disturbance propagated through the ether with the velocity of light. Some recent measurements by Marx<sup>1</sup> confirm this view, for he succeeded in showing that they do, in fact, travel with this velocity. If the  $\gamma$  rays were of this nature their behaviour would be explained, and on the whole the balance of evidence supports this view rather than the corpuscular hypothesis. The discussion as to the nature of Röntgen and  $\gamma$  rays has quite recently been revived by Bragg in a paper in the *Philosophical Magazine* (October, 1907). The view is there expressed that these rays may consist of uncharged material particles formed by the combination of an  $\alpha$  and  $\beta$  particle. It is shown that such a doublet would exhibit the properties of the  $\gamma$  rays.

<sup>1</sup> Marx, *Annalen der Physik*, Vol. XX, p. 677. 1906.

## CHAPTER V

### PHYSICAL AND CHEMICAL EFFECTS OF THE RADIATIONS FROM RADIOACTIVE BODIES

THE most important properties of the various radiations emitted by radioactive bodies have been carefully studied in the last chapter. There still remain to be considered certain physical and chemical effects which are produced when the radiations from a radioactive substance are allowed to fall upon certain bodies.

#### FLUORESCENCE PRODUCED BY RADIUM

We have already seen that when the radiations from radium fall upon certain substances a brilliant fluorescence is produced. Thus if a screen of zinc sulphide is interposed in the path of the rays, the screen can be seen in the dark on account of the fluorescence which has been induced in it. This action has already been studied, and it has been shown that at

any rate when the fluorescence is due to the  $\alpha$  rays the effect may be resolved by means of the spinthariscopes into a large number of minute flashes produced whenever an  $\alpha$  particle strikes the zinc sulphide.

The property is a fairly general one, and is not confined to zinc sulphide. Thus platino-cyanides which we have seen fluoresce under the influence of the Röntgen rays may also be excited by the radiations emitted by radium. Furthermore, many crystals fluoresce brilliantly under the action of the radiations; thus, for example, the diamond and the ruby exhibit this property to a marked extent, as do crystals of fluorspar, and to a less extent certain kinds of glass.

With this phenomenon of fluorescence there are often associated certain secondary effects in the bodies which are caused to fluoresce. Thus it is frequently noticed that when a body has been excited for some time to fluorescence by the action of the rays from radium its colour changes. For instance, if ordinary soda glass is exposed to the radiations from radium the glass gradually as-

sumes a beautiful violet colour. Similarly if potash glass is treated in the same way, the glass which previously was colourless is now found to possess a distinct brown coloration. It is on this account that the glass vessels used to contain radium salts or their solutions are nearly always found to be more or less highly coloured. By strongly heating the glass the coloration may be removed and the glass restored to its original condition.

Similar fluorescent and colour effects can be advantageously observed and studied in the case of crystals of fluorspar. This substance occurs in nature in many different varieties. In some cases the crystals are found quite colourless and transparent; in others the crystals are of various tints, which may be blue, green, yellow, or violet. If such crystals are exposed to the action of the rays from radium they are seen to fluoresce brilliantly, and the fluorescence persists for a considerable time after the removal of the radium. In addition to this action the colour of the crystal is frequently changed; thus a colourless crystal may become coloured, or the tint

of a crystal already coloured may be intensified. Now it is well known that if a coloured crystal of fluorspar is heated it is thereby discoloured, and the effect of subjecting a crystal which has been so treated to the action of the rays from radium is of interest.

There is a particular variety of fluorspar known as "blue-john," which occurs in Derbyshire, coloured so deeply violet that a crystal of any considerable size appears almost black. If such a crystal is strongly but carefully heated, it is deprived of its colour and becomes perfectly transparent. On now exposing the crystal to the action of the rays from radium, the crystal becomes again coloured, but there is a remarkable fact about the experiment, namely, that the crystal does not assume its original violet colour but is transformed into a crystal of sky-blue tint. At the same time the crystal has become strongly fluorescent, and may be seen in a dark room by the light which it emits. This fluorescence persists for a remarkably long time, the duration of which may be gathered from the following experiment. Four crystals of different samples of

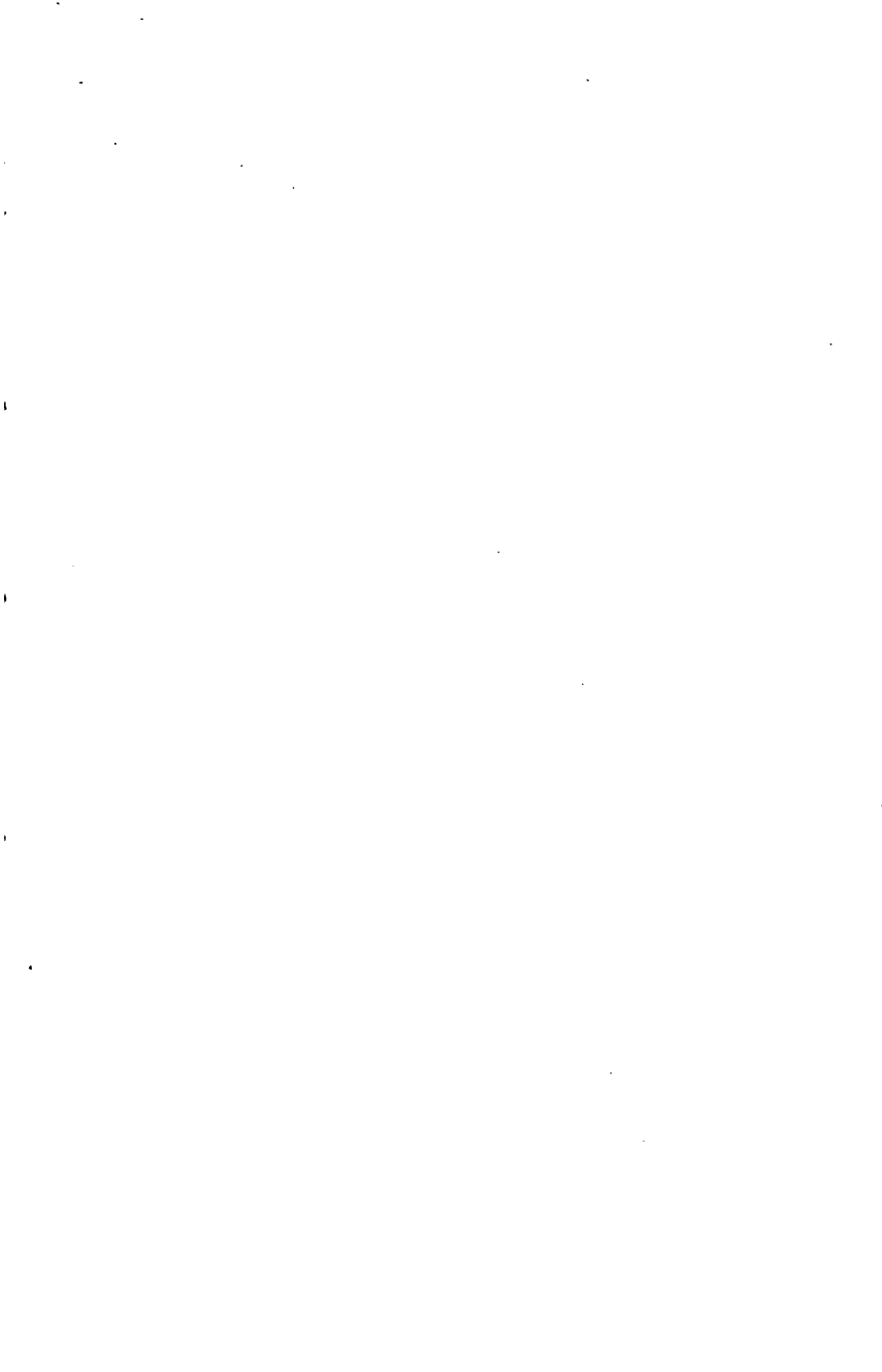




FIG. 21.—Photograph of crystals of fluor-spar after exposure to radium radiations.



FIG. 22.—Photograph of crystals in Fig. 21 ten days after removal from influence of radium.



fluorspar were exposed to the  $\beta$  and  $\gamma$  radiation of a considerable quantity of radium for some days, after which they were removed and found to have been coloured by the radiation which had been falling upon them. The crystals were then placed upon a sensitive photographic plate at its four corners and left to stand for an hour in a dark room. As might have been expected, on developing, the plate was found to have been affected by the fluorescent crystals which had been placed upon it. This is shown in Fig. 21. The experiment was repeated at various intervals, and even after a fortnight three of the four crystals were able to affect the photographic plate when the latter was exposed to their action for an hour. The action of the fourth crystal had, however, by that time become too feeble to be detected by the photographic plate. The result obtained, ten days after removal from the radium, can be seen from Fig. 22. It will be noticed that although the intensity of the impression obtained was much less than immediately after removing the crystals from the action of the radium, the effect is still appreciable.

It might at first sight seem that the crystals had in some way acquired the property of emitting some radiation other than light after exposure to the radium. But if this is so, the rays emitted must be very easily absorbed, for by interposing a thin sheet of black paper between the crystals and the photographic plate the action can be completely stopped. On the other hand this is just what would be expected if the action were due to the light emitted by the crystals.

#### PHOSPHORESCENCE OF RADIUM COMPOUNDS

There is an interesting property of radium salts which is allied to those which have just been discussed. When a compound of radium is examined in a dark room it is found that it emits a light which can easily be seen from considerable distances if the room is sufficiently dark.

In this connection the somewhat remarkable observation was made by the Curies that radium which contained a certain amount of barium salt mixed with it was frequently more luminous than purer specimens of radium salts. Now, remembering the power of pro-

ducing fluorescence which radium exerts upon many crystalline bodies, this fact, which is at first sight remarkable, can be explained. The barium salt mixed with the radium is constantly exposed to the radiations which the latter emits, and consequently becomes fluorescent; but when the radium is still further purified there is no longer any barium salt left to be caused to fluoresce, and as it so happens that radium salts themselves fluoresce less under the action of the rays than barium salts, the light emitted by a purer sample of radium salt is fainter than when in a state of less perfect purification.

There are other facts connected with the self-luminosity of radium salts which are worthy of brief mention. The presence of moisture appears to diminish the luminosity of radium salts, but if the moisture be carefully removed the original luminosity is regained. Again, with the lapse of time radium salts seem to gradually lose their power of emitting light. The power can, however, be restored to the salts by recrystallizing from a solution of water.

## CONDUCTIVITY PRODUCED IN DIELECTRICS

The conductivity produced in gases by the rays from radioactive bodies has been noticed and somewhat fully considered in chapter I. The power which the rays have of rendering conductors of electricity bodies which are normally insulators is not confined to matter in the gaseous state, and similar effects may frequently be produced in liquids and solids. This interesting fact was discovered by P. Curie<sup>1</sup> who showed that the current passing between two concentric cylinders immersed in a liquid insulator was considerably increased by the presence of radium. Similar effects were also noticed with solids.

The subject has since been carefully investigated by Jaffé<sup>2</sup> for the case of petroleum ether. The subject is complicated by the fact that the effect of radiation on petroleum ether varies with the specimen, and it was found that the less conducting the ether was when not subjected to any radiations the more sensitive it was to their influence. Moreover

<sup>1</sup> P. Curie, *C. R.*, C XXXIV, p. 420. 1902.

<sup>2</sup> G. Jaffé, *Journal de Physique*, April, 1906.

the conductivity of any given specimen of petroleum ether did not remain constant but increased gradually with time if a current of electricity were passed through it. In one case a change of as much as 13 per cent was observed after the current had passed for thirteen days.

In spite of these difficulties Jaffé was able to show that the laws governing the conductivity of petroleum ether were similar to those for gases, except that it was impossible to reach the saturation point when, as it will be remembered, the current is independent of the difference of potential. The fact is no doubt due to the greater resistance which the ions experience when moving through a liquid, so that enormous fields are necessary to drag them to the electrodes before recombination has taken place.

#### CHEMICAL EFFECTS OF THE RAYS

We have already noticed several cases of chemical action produced by the rays from radioactive substances. Thus it was pointed out that one of the methods of detecting and

studying radiations consisted in observing the effects produced on photographic plates, which is, of course, an example of chemical action ; for, as is well known, such action is due to chemical changes produced in the silver chloride contained in the film upon the plate.

As another instance of chemical action which has already been considered, may be mentioned changes of colour produced in glass and in certain crystals. The nature of the changes involved in these colour transformations has received some attention, but though they seem to be due to some chemical change induced by the rays, the nature of the change is not fully understood. It has been suggested that this action on glass is brought about by the separation of the alkali metals in the glass which then form a solid solution, but it is questionable whether this explanation is correct and the view mentioned awaits further confirmation. There are many other instances of chemical action produced by the radiations from radium, and it would be useless to attempt to enumerate even the most important. But there are certain reactions which

should be mentioned in passing because they are constantly occurring when manipulations of radium salts are undertaken. Thus when a radium salt is dissolved in water it is found that the water is gradually split up into its constituents, oxygen and hydrogen, and consequently the air in contact with such a solution always contains some free hydrogen. Again oxygen is converted into ozone by the radiations, and the presence of this substance may always be detected in the neighbourhood of a radium salt by its peculiar odour.

## CHAPTER VI

### THE EMANATIONS

#### THE DISCOVERY OF THORIUM EMANATION

ATTENTION has already been called to the fact that the current between two plates of a condenser, one of which is covered with a thorium compound, varies in a somewhat capricious manner. The phenomenon was studied by Owens,<sup>1</sup> who showed that the variations could be traced to air currents, and that, if care were taken to avoid this disturbing cause, constant results could be obtained for the current passing through the gas emitted by a definite quantity of thorium.

It remained for Rutherford<sup>2</sup> to give an explanation of the manner in which air currents disturbed the observations. If an air current is passed over a thorium compound,

<sup>1</sup> Owens, *Phil. Mag.*, p. 360, Oct., 1899.

<sup>2</sup> Rutherford, *Phil. Mag.*, p. 1, Jan., 1900.



or, what is more satisfactory, if air is drawn through a solution of thorium nitrate in water, this air is found to have temporarily acquired the property of discharging an electrified body. Now this in itself is not surprising, for we know that an ionized gas retains its ionization for a finite time, and we know that the gas must have been in this condition on leaving the thorium solution. The remarkable fact about the experiment was that the conductivity of the air could not be removed by any of the ordinary methods for withdrawing ions from an ionized gas, *i.e.* filtration through cotton wool, bubbling the gas through water contained in a wash bottle, or passing it through a strong electric field. It is therefore clear that something is taken up by the air in its passage through the thorium solution which is capable of ionizing the gas.

Rutherford made a number of experiments to find out whether some solid particles of matter were withdrawn from the thorium solution and carried along with the air current. The fact that the air could be filtered through

cotton wool was antagonistic to the view that the air stream contained any solid particles, and a multitude of evidence pointed to this view being incorrect. The nature of the substance which was withdrawn from the thorium was for some time not understood, and Rutherford proposed the term *emanation* for this substance. The emanation has since been demonstrated to consist of a gas, but the original name has been preserved, although it is perhaps no longer very appropriate. Attempts have been made to substitute other names more suggestive of the gaseous nature of the emanation, but there seems no particular object for the change, and the original name has at least the merit of historical interest.

#### EMANATIONS FROM RADIUM AND ACTINIUM

The discovery of the thorium emanation was so remarkable that scientists set to work to find out whether other radioactive substances gave off similar emanations, and, in fact, to ascertain whether this property was one generally exhibited by radioactive bodies.

The search was rewarded by the discovery of two new radioactive gases. Dorn<sup>1</sup> found that radium gave off a gas similar to the thorium emanation, and Debierne<sup>2</sup> discovered that actinium behaved in the same way. No emanation has yet been obtained from polonium; and though repeated efforts have been made to demonstrate the existence of an emanation from uranium, all attempts in this direction have hitherto resulted in failure.

It appears, therefore, that at least three radioactive substances exhibit the power of emitting emanations, but the property does not seem to be shared by all the radioactive elements, and cannot therefore be regarded as a phenomenon necessarily associated with radioactive processes.

The properties of the emanations from thorium, radium, and actinium are in most respects very similar, and we will therefore concentrate our attention on the radium emanation, which has been most carefully

<sup>1</sup> Dorn, *Abhandlungen de Naturforscherversammlung zu Halle*. 1900.

<sup>2</sup> Debierne, *Comptes Rendus*, CXXXVI, p. 146, 1903.

studied, merely pointing out the important differences between it and its analogues thorium and actinium.

#### PREPARATION OF RADIUM EMANATION

The amount of emanation obtained by passing an air current over a radium salt is considerably influenced by the physical state of the radium salt. For instance, it is found that if the radium salt is very carefully dried, the amount of emanation taken up by a stream of dry air passing over it may be reduced to about one-fifth of the amount taken up when the salt has been exposed to damp air for some time. If, on the other hand, the radium salt is dissolved in water and a current of air passed through it, the amount of emanation given up to the air is at first enormously increased; if, however, the air current is continued for some time, the radium is almost completely deprived of its emanation, and when in this condition is said to be *de-emanated*. When large quantities of emanation are required this method of preparation is frequently

adopted, but the same result may often conveniently be obtained by strongly heating the radium salt. We see, then, that only a definite quantity of emanation can be obtained from any given quantity of radium at any time, but it is found that if the radium is allowed to stand, it gradually regains its power of giving off the emanation. At the end of one or two weeks nearly the maximum quantity of emanation may again be withdrawn from it.

#### RATE OF DECAY OF THE EMANATION

If some radium emanation is enclosed in a gas-tight vessel, then it is found that the ionizing power of the gas gradually diminishes with time, the rate of decrease of the intensity of radiation being always proportional to the quantity present. Expressed in mathematical symbols we have, if  $I$  represents the intensity of the radiation at any moment and  $t$  the time,

$$-\frac{dI}{dt} = \lambda I \quad (1)$$

where  $\lambda$  is a constant.  $dI \cdot dt$

By a very simple integration we have from (1) that

$$- \lambda t = \log_e I + C$$

where  $C$  is a constant.

Now if  $I_0$  is the intensity of radiation when  $t = 0$

$$C = - \log_e I_0$$

whence 
$$- \lambda t = \log_e \frac{I}{I_0}$$

or 
$$I = I_0 e^{-\lambda t} \quad (2)$$

which gives the rate at which the activity of the emanation disappears.

Now this equation is of great importance because it is found that all known radioactive substances, of which we shall see there are a great number, have been shown to lose their radioactive property according to this law, except uranium, radium, and actinium, the radio-activity of which seems to be independent of time. There is, however, much indirect evidence to show that even these three bodies form no exception to this law, their rate of decay being merely too slow to admit of direct observation within a reasonable time.

The constant  $\lambda$  differs for different radioactive bodies, being different, for instance, for the emanations from radium and thorium and again different for the emanation from actinium. It seems, however, to be unchanged for one and the same radioactive product when subjected to very varied chemical or physical treatment; hence the measurement of this constant often affords a trustworthy means of identifying a radioactive substance. Thus radium emanation may be heated or cooled over very wide limits, and subjected to high pressures or to the action of any known chemical reagent, without producing any change in the rate at which it loses its activity. In fact, it has been thought that this rate of decay is independent of any agent to which it can be exposed by man. Doubt has, however, recently been thrown on this supposition by some experiments by the author,<sup>1</sup> in which it is shown that a temperature of about 1100° centigrade may, under suitable conditions, produce a small change in the activity of radium emanation. We shall have to re-

<sup>1</sup> Makower, *Proc. R. S., A.*, Vol. LXXVII, p. 241, 1906.

turn to this interesting question more fully later.

To return to the question of the disappearance of the emanation when separated from radium. It has been mentioned that the emanation gradually loses its radio-activity at a definite rate, which we may consider to be independent of the physical conditions to which it is subjected, and that this rate of loss of activity, or, as it is more often called, the decay of activity, can be represented by the constant  $\lambda$  defined above. There is another way of defining the rate of decay which is generally more convenient. It is usual to express this quantity by the time taken for the activity of the emanation to fall to half its original value; this time is, of course, independent of the initial value, and is therefore a convenient measure of the rate of decay.

#### MEASUREMENTS OF THE RATES OF DECAY OF THE EMANATIONS

Measurements of the rates of decay of the emanations have been carefully made by various investigators.



The rate of decay of radium emanation was first studied by Curie.<sup>1</sup> Some radium emanation was hermetically sealed in a glass tube and its activity measured at frequent intervals for several days, and it was found to have lost half its activity in 3.99 days.

Using a somewhat different method, Rutherford and Soddy<sup>2</sup> found the value 3.71 days. These two values differ by nearly 10 per cent., which is certainly more than can be accounted for by experimental error, and it would be interesting to know what is the cause of this discrepancy. Rutherford and Soddy measured the activity of the emanation by the intensity of the  $\alpha$  radiation emitted by it, whereas in Curie's experiment the  $\beta$  and  $\gamma$  rays were used, which we shall see later are not given out by the emanation itself at all, but by an active deposit which the emanation precipitates on the walls of the vessel in which it is contained, and it is probably to this circumstance that the discrepancy is due. There can, however, be little doubt that the value obtained by

<sup>1</sup> Curie, *C. R.*, CXXXV, p. 857, 1902.

<sup>2</sup> Rutherford and Soddy, *Phil. Mag.*, April, 1903.

Curie is considerably too high, although that obtained by Rutherford and Soddy is probably slightly too low. A careful determination of the rate of decay of radium emanation recently carried out by Rümelin gave the value 3.75 days for the time to fall to half value.<sup>1</sup>

The most accurate determination of the rate of decay of thorium emanation has been made by Le Rossignol and Gimmingham,<sup>2</sup> who find that this emanation decays to half value in fifty-one seconds. The emanation from actinium decays even more quickly, falling to half value in 3.9 seconds.<sup>3</sup>

#### RELATION BETWEEN THE DECAY OF RADIUM EMANATION AND THE RECOVERY OF RADIUM AFTER DE-EMANATION

After radium has been de-emanated it is found that it gradually regains its power of emitting emanation. After de-emanation the radium gives off far less radiation than when in

<sup>1</sup> *Phil. Mag.*, Oct., 1907.

<sup>2</sup> Le Rossignol and Gimmingham, *Phil. Mag.*, July, 1904.

<sup>3</sup> Debierne, *Comptes Rendus*, CXXXVI, p. 146, 1903.

the normal condition, its activity having fallen to about one-quarter of its initial value. It therefore seems that by the withdrawal of the emanation, which is itself radioactive, the activity of the radium has been reduced to one-quarter, so that three-quarters of the radiation emitted by radium under normal conditions comes not from the radium itself but from the emanation contained in it.

This fact is further confirmed by measurements of the activity of the emanation and the radium at definite times after their separations. It is found that as the radium gradually regains its property of giving off emanation when heated, its power of giving off rays increases, and it regains half its lost activity in about four days. Now we have seen that the emanation loses half its activity in the same time, so that there is evidently some close agreement between these two phenomena. If now the activities of the radium and emanation respectively are measured at successive intervals after separating the emanation from the radium, it is found that *the sum of the activities of the two separated bodies is constant.*

This is shown in Fig. 23, in which curve A gives the decay of the emanation, curve B the rise in activity of the radium, and the straight line C, which is parallel to the axis of time, the sum of these two activities at successive times. Now this is a result of the greatest importance; for although it is possible to separate the emanation from the radium with which it is usually associated, the rates at which the emanation decays and the radium regains its power of emitting the emanation are exactly complementary, a fact which may be interpreted as meaning that with a definite quantity of radium there is always associated a definite quantity of emanation, no matter whether the emanation is in contact with the radium or not. This is precisely what might be expected if the emanation which we know to be constantly disappearing were being re-formed by the radium with which it is associated.

To explain these phenomena Rutherford and Soddy have suggested the interesting hypothesis, which is usually known as the "disintegration theory," that the radium atom de-

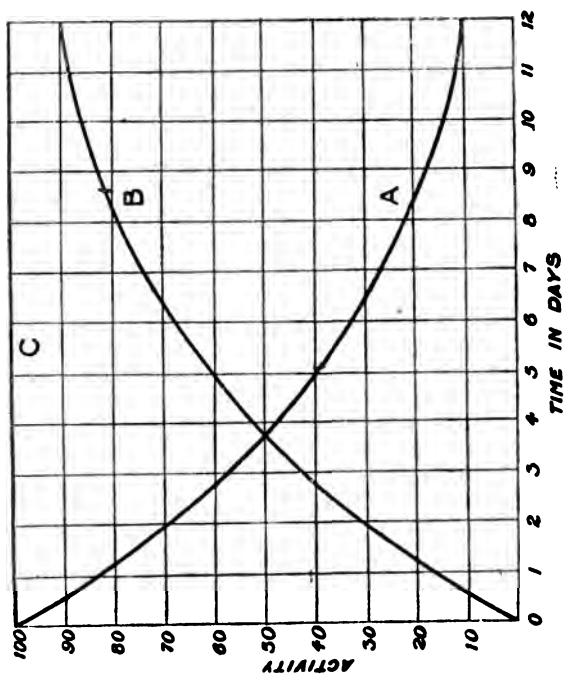


FIG. 23.—Decay and recovery curves of radium emanation.



composes into the emanation as it emits the  $\alpha$  rays, which we have seen are constantly given off. We shall see later that this theory is of very wide application and is of service in explaining many other radioactive processes. An obvious objection to this theory is that, if this view is correct the radium must be disappearing as the emanation is formed, and eventually we should find the quantity of radium diminished, and this is contrary to experience, for no loss of weight can be detected even when radium is kept for years. The objection can, however, be directly met by calculation, whereby it can be shown that to supply the minute quantity of emanation which is evolved by the radium it would be necessary for only a quite unmeasurably small quantity of radium to be decomposed during the time of any experiment hitherto performed.

This theory has been the subject of much adverse criticism, but is now almost universally accepted as correct. A discussion of all the evidence which points to this conclusion must, however, be postponed to a later stage.

CHEMICAL AND PHYSICAL PROPERTIES OF THE  
EMANATIONS

It is important to know the chemical and physical properties of these newly discovered gases, and much has been done towards this end. But the investigation is fraught with unusual difficulty, for it is impossible to obtain more than the minutest quantities of the emanations, so that most of the manipulations used in experimenting with gases become impossible to perform. In a very beautiful experiment performed by Ramsay and Soddy<sup>1</sup> it was shown that from 60 milligrams of radium bromide it was possible to collect in a fine capillary tube a measurable quantity of the emanation.<sup>2</sup> The gas could be easily seen in the dark by the phosphorescence produced in the walls of the capillary tube, and the volume of the gas decreased during the course of a few days in a manner such as might have been

<sup>1</sup> Ramsay and Soddy, *Proc. Roy. Soc.*, LXXII, p. 204, 1903.

<sup>2</sup> In the earlier experiments it was estimated that one gramme of radium gave rise to about one cubic millimetre of the emanation. But later experiments by Cameron and Ramsay (*Jahrbuch für Radio-aktivität und Elektronik*, IV, 3, pp. 253-74) seem to show that the above estimate is too low.



expected from a knowledge that the activity of the emanation decays with time. In spite of the fact that only these very small quantities of emanation can be isolated, and that it is difficult to separate so small a quantity of gas from air and other impurities with which the emanation is always mixed when given off by the radium, it has been found possible to investigate many of the properties of the emanation.

Both radium and thorium emanations have been exposed by Rutherford and Soddy<sup>1</sup> to a large variety of chemical reagents under different conditions to ascertain the chemical behaviour of these newly discovered gases. The emanation was carried over the different reagents by a stream of gas, itself inert to the reagent used, and the activity tested before and after this treatment. No chemical action could, however, be detected between the emanations and any of the reagents used. Acids, alkalis, red-hot platinum black, lead chromate, magnesium powder, palladium black, and zinc dust alike failed to produce any

<sup>1</sup> Rutherford and Soddy, *Phil. Mag.*, November, 1902.

effect on the emanation when passed over them; in fact, these new gases seemed to be chemically quite inert. Ramsay and Soddy<sup>1</sup> have since sparked a mixture of the emanation with oxygen in the presence of alkali and heated it in the presence of magnesia lime, but even this drastic treatment left the emanation unaltered. Thus these gases are unlike any known form of matter except the members of the argon group of elements, with which it is therefore usual to classify them. It might, therefore, be expected that their atomic weights would be such as to make them fall into the argon group of elements in Mendeléeff's periodic table of atomic weights. It was therefore a matter of some importance to endeavour to determine the atomic weights of the emanations. The problem is, however, fraught with the usual difficulty in dealing with these bodies, namely, that it is impossible to obtain them in sufficient quantity to employ the usual methods for atomic weight determinations. It would, for example, be

<sup>1</sup> Ramsay and Soddy, *Proc. Roy. Soc.*, LXXII, p. 204, 1903.

quite hopeless to attempt to determine the density of the emanations by direct experiment. There is, however, an indirect method of measuring the density of gases which can be applied to the present case with moderate accuracy.

#### DIFFUSION OF THE EMANATIONS

It is well known that if two dissimilar gases are introduced into different parts of the same enclosure they will gradually mix, even though they are not subjected to any mechanical disturbance. The molecules of which the gases are composed are in constant motion, and they mix with each other or, as it is more usually expressed, *diffuse* through each other, and after sufficient time has been allowed for equilibrium to be established, the molecules of both gases are found to be uniformly distributed throughout the enclosure. The rate at which this process takes place varies greatly, however, with the chemical nature of the gas used. If, for instance, we take any one gas, say air, and allow first hydrogen and then carbon dioxide to diffuse

into it, we find that the diffusion takes place very much faster in the first case than in the second, indicating that the hydrogen molecules move faster through the air than carbon dioxide molecules. Quantitative experiments have been made by Graham, Loschmidt, and others, who have shown that the rate of diffusion of a gas depends on its molecular weight being greater for gases whose density is small, and smaller for those gases whose density is great. The rate of diffusion is found approximately to follow a very simple law first enunciated by Graham, according to which *the rate of diffusion of a gas into any other gas is inversely proportional to the square root of its density.*

It will thus be seen that from measurements of the rate of diffusion of one gas into another its density may be calculated, and consequently the molecular weight of a gas may be determined in this way.

The first attempt to apply this method was made by Rutherford and Brooks.<sup>1</sup> The ex-

<sup>1</sup> Rutherford and Brooks, *Trans. Roy. Soc., Canada*, 1901 ; *Chem. News*, 1902.

periment consisted essentially in filling one portion of a long cylindrical vessel with air containing some radium emanation and, by opening a shutter provided for the purpose, allowing the emanation to diffuse into another portion of the vessel in which the conductivity of the air caused by the emanation diffusing into it could be measured at definite times after the beginning of the experiment. In this way a measure was obtained of the rate of diffusion of the emanation through air.

The experiment is, however, subject to extremely great difficulties, because even small differences of temperature in different parts of the diffusion vessel cause air currents to be set up which make the diffusion appear to take place too quickly. Allowing for all disturbing causes, the experiments indicated that the molecular weight of the emanation was about 100, the atomic weight of hydrogen being taken as unity.

Curie and Danne,<sup>1</sup> by measuring the rate of diffusion of radium emanation through capillary tubes, arrived at a somewhat lower value

<sup>1</sup> Curie and Danne, *C. R.*, CXXXVI, p. 1314, 1903.

for the molecular weight; but no special precautions seem to have been taken to maintain constant temperatures, and the results obtained must therefore be regarded as only approximate.

To avoid the errors mentioned above and due to the production of air currents by local heating of the diffusion apparatus, the author<sup>1</sup> carried out a series of measurements of the rate of diffusion of radium emanation through plugs of various porous substances and compared this rate with that of well-known gases through the same plugs.

The apparatus used is shown in Fig. 24.

The air containing the emanation the rate of diffusion of which was to be determined, was contained in the "diffusion vessel" A, which was graduated downwards in cubic centimetres, the upper end being closed by a porous plug P.

To carry out an experiment a certain volume of air mixed with emanation was sucked into A and a known volume transferred through the stop-cock T into the vessel V, in

<sup>1</sup> W. Makower, *Phil. Mag.*, Jan., 1905.

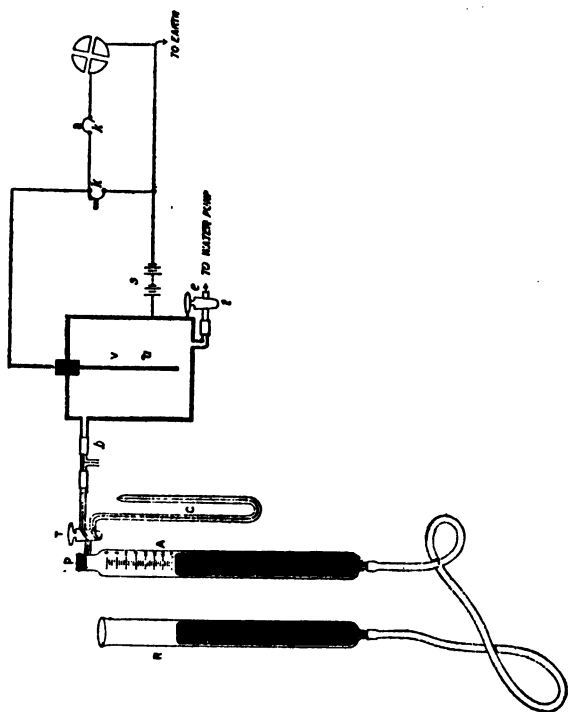


FIG. 24.—Apparatus for determining molecular weight of radium emanation.





which the conductivity of the air could be measured. The emanation was then allowed to diffuse through the porous plug for a known time, when another sample was transferred from A into V. A measure of the relative concentrations of the emanation in A at the beginning and end of the experiment was thus obtained, which gave a measure of the rate at which the emanation diffused through the porous plug.

By experimenting with gases of known density, such as oxygen, carbon dioxide, and sulphur dioxide, it was possible by chemical analysis to determine the rates of diffusion of these gases through the same plug; by comparing these with the rates of diffusion of radium emanation and applying Graham's Law connecting the density of a gas with its rate of diffusion the density of the radium emanation was deduced.

The molecular weight was found in different experiments to vary from 85.5 to 99, values which do not differ greatly from that given by Rutherford and Brooks. A further series of experiments showed that the molecular

weight of thorium emanation does not differ much from this value, a conclusion which has also been reached by Rutherford by an entirely different method.

It is interesting to speculate as to the position of the emanation in the periodic table of elements. If the above described determinations of the molecular weight may be accurately trusted, then the emanations would, on the assumption that they are monatomic gases, fall in the vacancy in the fluorine group between Molybdenum ( $\text{Mo}=96.0$ ) and Ruthenium ( $\text{Ru}=102$ ). But we have seen that the emanations are chemically quite unlike any of the members of the fluorine group, and that they resemble most clearly the members of the helium group of elements. If we may assume that the atomic weights obtained for the emanations are somewhat too low, it is possible to group them in the helium group between Krypton ( $\text{Kr}=82$ ) and Xenon ( $\text{Xe}=128$ ). There is, however, even in this case the difficulty of finding a place both for radium and thorium emanations, for which, as we have seen, the atomic weights are nearly

equal ; it seems therefore doubtful whether it will prove at all possible to classify these new bodies in the periodic system.

#### CONDENSATION OF THE EMANATIONS

There seems to be little doubt that the emanations from radium and thorium consist of minute quantities of gaseous substance given off by these two radioactive elements. We have already considered much of the evidence which supports this view, and we have seen that the emanations may be manipulated just as any ordinary gas, and that their chemical and physical properties may be studied by modification of the methods used when dealing with other gases.

There is yet another property of the emanations which lends support to this view. There is only one gas—helium—which is known in the gaseous state only, in spite of the repeated attempts which have been made to convert it to a liquid. All other gases may be converted to the liquid or solid state by sufficiently reducing their temperature. It might therefore be expected that the

emanations, if gaseous at ordinary temperatures, should be condensed to the liquid or solid state by a suitable reduction of temperature. This was, in fact, found to be the case by Rutherford and Soddy,<sup>1</sup> who have made a careful study of the matter.

The emanations from radium and thorium respectively were passed in a slow stream of air or other gas through glass spirals immersed in liquid air by means of which a temperature of about  $-190^{\circ}$  C. may be obtained. It was found, by testing the gases which passed out through the spirals, that both in the case of radium and thorium the emanation remained in the cooled spiral. In the case of the radium emanation, the rate of decay of which is slow, it was possible to recover the emanation by removing the spiral from the liquid air and allowing the temperature to rise; but with thorium emanation the rate of decay is so rapid that the manipulations cannot be carried out sufficiently quickly for the purpose.

The boiling point of radium emanation was

<sup>1</sup> Rutherford and Soddy, *Phil. Mag.*, May, 1903.

found to be  $-150^{\circ}\text{C.}$ , whereas that of thorium emanation was  $-120^{\circ}\text{C.}$  How nearly these values may be considered as the true boiling points is a matter of some doubt, for the emanations were always mixed with some other gas so that their partial pressure was exceedingly small. Under these conditions the molecules of the emanation are separated from each other by comparatively great distances, a circumstance which would prevent the emanations from condensing as readily as if they were at atmospheric pressure. The values obtained are, however, probably a fair approximation to the truth.

## CHAPTER VII

### THE ACTIVE DEPOSITS PRODUCED BY THE EMANATIONS

#### INTRODUCTION

A MOST remarkable discovery was made by M. and Mme. Curie<sup>1</sup> during their investigation of the properties of radium. It was found by them that bodies left for some time in the presence of radium salts themselves became temporarily radioactive, having acquired the power of emitting radiations even when removed from the presence of the radium. By their proximity to the radium the bodies in question had become possessed of the radioactive properties of the radium itself, and the bodies were said to have had radio-activity *induced* or *excited* in them. Soon after this discovery Rutherford<sup>2</sup> showed that similar

<sup>1</sup> M. and Mme. Curie, *C. R.*, CXXIX, p. 714, 1899.

<sup>2</sup> Rutherford, *Phil. Mag.*, Jan. and Feb., 1900.

phenomena were exhibited by bodies exposed to thorium, and Debierne<sup>1</sup> found that actinium could induce radio-activity on surrounding bodies to a marked extent.

The phenomena were at first supposed to be due to some peculiar influence exercised on a body by the radiation from the radioactive substance falling on it, and the question arose, whether, by the absorption of the radiation falling upon it, the body had not suffered some subtle change whereby it had for a short time acquired the property of emitting radiations. This view, however, soon proved to be untenable, for bodies situated in a closed vessel containing some radium salt acquire the property equally, either when exposed to the direct radiation from the radium or when carefully screened from it.

When a solution of a radium salt was suspended inside an enclosure in a glass vessel open at the top any other body placed inside the enclosure became active, nor did the quantity of activity induced on the body depend in any way on the material of which

<sup>1</sup> Debierne, *C. R.*, CXXXVIII, p. 411, 1904.

it was composed. The activity was, however, proportional to the area exposed, but was unaffected by the interposition of a lead screen between the body and the radium so as to cut off the direct radiation from the latter.

The experiment was next tried of closing the glass vessel containing the radium, with the remarkable result that no trace of induced activity could be detected on bodies situated in the larger vessel. Now this experiment indicates that to produce the phenomena under consideration it is necessary for some material substance to pass from the radium to the body on which the activity is to be induced, and that by preventing this transference we can shield the bodies from the influence of the radium. Combining this conclusion with the fact that it is only those bodies—radium, thorium, and actinium—which give off an emanation, which have the power of inducing activity on neighbouring bodies, it was not unreasonable to attribute the observed effect to the direct action of the emanation.



PRODUCTION OF INDUCED ACTIVITY BY THE  
EMANATION

The view that the emanation is directly responsible for the production of induced activity was first expressed by Rutherford, who pointed out that this hypothesis was capable of explaining all the facts known at the time. Subsequent discoveries have left no doubt as to the close relation existing between the emanation and induced activity. The experiments upon which this theory was based were first made with thorium, which, as we have already seen, is similar to radium in its power of producing induced activity on neighbouring bodies. If, on the one hand, the emanation was prevented from reaching bodies near the thorium salt they remained quite inactive; but, on the other hand, if steps were taken to insure the emanation coming into contact with a body, the induced activity at once began to appear on it, although the emanation had been removed from the thorium and carried by a stream of air beyond its influence. Quantitative

measurements on the magnitude of the induced activity showed that the activity communicated to a body by the emanation at first increased with the time for which it had been exposed to the emanation and ultimately became constant. Moreover, the magnitude of this constant value was, at least, roughly proportional to the amount of emanation to which it had been exposed when both these quantities were measured by the ionization which they could produce in gases.

#### NATURE OF THE INDUCED ACTIVITY.

The origin of the induced activity having thus been fixed, it was necessary to inquire more closely into the cause of the phenomena which have just been described. For though it be admitted that bodies are affected by radium, thorium, and actinium only by the intervention of the emanations which they emit, yet we are no nearer to an understanding of the changes which have taken place in the activated body.

A number of experiments by Von Lerch,<sup>1</sup>

<sup>1</sup> Von Lerch, *Ann. d. Phys.*, XII, 745, 1903.

Rutherford,<sup>1</sup> and others have greatly extended our knowledge with regard to this matter.

It can readily be shown that the activity induced on a body is confined to the surface layers of the substance, by the removal of which the substance may be brought back into an inactive state. If, for instance, the surface of an active metallic plate is removed by rubbing with emery paper, it will be found that the plate has lost its activity; or, again, if the surface of an active copper plate is removed by dissolving it in nitric acid, the interior of the plate does not display any sensible activity. If the solution thus obtained is evaporated to dryness and the residue examined, the activity is again recovered, showing that by dissolving the surface the activity has been transferred from the plate to the solution.

A fact which is, however, perhaps more striking still is that it is not even necessary to dissolve away the surface of the active plate; the activity may also be removed by

<sup>1</sup> Rutherford, *Phys. Zeit.*, III, 245, 1902.

reagents which have no chemical action on the metal of which the plate is composed.

To demonstrate this, a platinum surface was rendered active by exposure to the emanation and then immersed in various solvents. Organic solvents such as alcohol and ether were without effect on the platinum, which retained its activity after treatment with these liquids. But when the platinum had been immersed in a solution of hydrochloric acid it no longer possessed any radioactive properties but the solution of acid had become strongly active. The activity of the platinum could thus be removed by mechanical and chemical means just as if it were due to a thin superficial film of a body with definite chemical properties.

Now we have already learnt that the emanations consist of material particles in the gaseous form which are constantly undergoing some change by which they sacrifice their radioactive properties. It follows, therefore, that unless there is an actual destruction of matter, the matter which goes to constitute the emanation must appear in some form

inside a vessel containing the emanation after it has lost its radioactive characteristics. When it is remembered that the emanations exist only in such quantities that their detection is almost impossible except on account of their radio-activity, it might easily happen that the matter left over after the activity of the emanation has decayed would escape detection, *unless it in turn itself displayed radio-activity*. In this case, however, there might be no more difficulty in detecting the matter produced by the emanation than the emanation itself. Rutherford has suggested that the presence of the induced activity on bodies exposed to the emanations may be accounted for in this way. The emanation which is constantly decaying is supposed to give rise to a substance deposited on bodies in its neighbourhood which itself decays with time, giving out radiations and accounting for the phenomena of induced activity. On this view, then, the induced activity is due to a deposit of material radioactive particles on bodies exposed to the emanation, which for brevity is usually

spoken of as the *active deposit* from radium, thorium, or actinium, as the case may be. We have already seen that emanations are constantly being formed from radium, thorium, and actinium, and it was pointed out that these bodies are regarded as decomposing into the emanations whilst at the same time emitting radiations. We see now that the emanations themselves give rise to radioactive deposits upon bodies with which they come in contact and that applying the same hypothesis as that adopted for the formation of the emanation, the active deposits may be considered as being formed by the decomposition of the emanations. We have thus a second case of an apparent transformation of one form of matter into another.

#### CHARGE CARRIED BY THE ACTIVE DEPOSITS

When an emanation is introduced into an enclosure we have seen that a uniform deposit of active matter is formed on the walls of the enclosure. The active matter appears to deposit itself equally on all parts of the enclosure, the activity of any portion of

the surface being proportional to its area. The uniformity of the distribution of the active matter may, however, be altered by means of an electric field.

If, for instance, an experiment is so arranged that one portion of the surface exposed to the emanation is maintained at a positive electrical potential, and another at a negative potential, then the whole of the active deposit (or as we shall see later, in the case of radium, *almost* the whole of the active deposit) is confined to the negatively charged portions in the field. This fact indicates that the particles which transmit the activity from the emanation to the walls of the containing vessel must be charged with positive electricity.

Now it has been shown by a variety of experiments, and notably by those of Battelli and Maccarrone,<sup>1</sup> who have made a very careful study of the matter, that the emanations are themselves uncharged. We are therefore driven to the conclusion that the

<sup>1</sup> Battelli and Maccarrone, *Phys. Zeit.*, VI., pp. 161-3, 1905.

active deposit acquires a positive charge during the process of its formation from the emanation or possibly subsequent to its separation from the emanation which produced it. The mechanism of the process involved is still not fully understood, and we shall return shortly to a consideration of this question on account of its theoretical importance. Apart, however, from any theoretical considerations, the fact itself is of the utmost practical utility in studying the properties of these radioactive deposits, for we have only to expose a negatively charged conductor, for example a wire, to the emanation in order to concentrate the maximum possible quantity of deposit on its surface. It is thus possible, without any difficulty, to obtain surfaces whose activity per unit area is enormously greater than could be obtained by any other means.

Turning to the theoretical aspect of the question, we have to inquire how it comes about that an atom of emanation which is itself uncharged can give rise to an atom positively charged with electricity. Remembering that the active deposit is produced from the



emanation by the expulsion of an  $\alpha$  particle which, we know, carries a positive charge, it is evident that if the  $\alpha$  particle is charged at the moment of its expulsion from the emanation, the residue which constitutes the active deposit should remain *negatively* charged, and this is the reverse of what is known to be the case. Doubt has, however, recently been thrown on the supposition that the  $\alpha$  particle is charged at the moment of its expulsion from the atom by Soddy,<sup>1</sup> who, as a result of experiments performed in very high vacua, claims to have proved that the  $\alpha$  particle acquires its charge by collision with air molecules after it has been expelled from the radioactive atom. But the experiment lacks confirmation, and as pointed out by Mr. Soddy<sup>2</sup> himself in a later communication, the deduction that the  $\alpha$  particle is uncharged at the moment of its formation cannot be taken as definitely established. Even adopting this view we should expect that the active deposit at the moment of its formation would be un-

<sup>1</sup> Soddy, *Nature*, August 2, 1906.

<sup>2</sup> *Ibid.*, March 7, 1907.

*charged*, but we are still as far as ever from explaining how the active deposit can become *positively charged*, which, as a direct result of experiment, we know to be the case.

THE MANNER IN WHICH THE ACTIVE DEPOSIT  
BECOMES CHARGED

Shortly after the discovery that the active deposit could be concentrated on a negatively charged body in an enclosure containing some emanation, it was shown by Rutherford<sup>1</sup> that, at least in the case of the thorium emanation studied by him, the concentration under consideration no longer took place if the pressure of the air with which the emanation was mixed were sufficiently reduced. In the experiments which he performed, he found that when the pressure of the air was reduced below one centimetre of mercury, less activity was concentrated on the negative electrode in an electric field than is the case under normal conditions. This experiment indicates that the air with which the emanation is mixed plays some important part in the acquisition

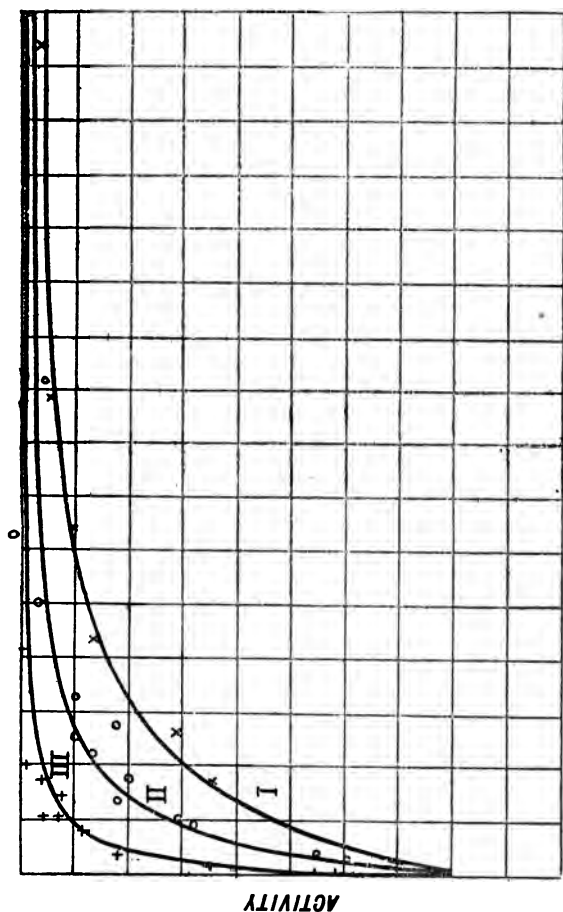
<sup>1</sup> Rutherford, *Phil. Mag.*, February, 1900.

of a positive charge by the active deposit which remains uncharged when the air is sufficiently attenuated. Some more recent experiments by the author<sup>1</sup> on the same subject show that similar results hold good for the radium emanation, and that the quantity of active deposit concentrated on the negative electrode at low pressures depends not only on the pressure of the air in the containing vessel, but also on the dimensions of the vessel used. The experiments consisted in exposing to the emanation from radium a metal rod fixed so as to be situated along the axis of a cylindrical metal vessel from which it was insulated. The rod was connected to the negative pole of a battery of cells, the vessel being connected to the positive pole. Matters were so arranged that the wire could be exposed to a constant quantity of radium emanation when mixed with air at different pressures. The quantity of active deposit concentrated on the rod was compared at different pressures, using vessels of various sizes, and it was found that the

<sup>1</sup> Makower, *Phil. Mag.*, November, 1905.

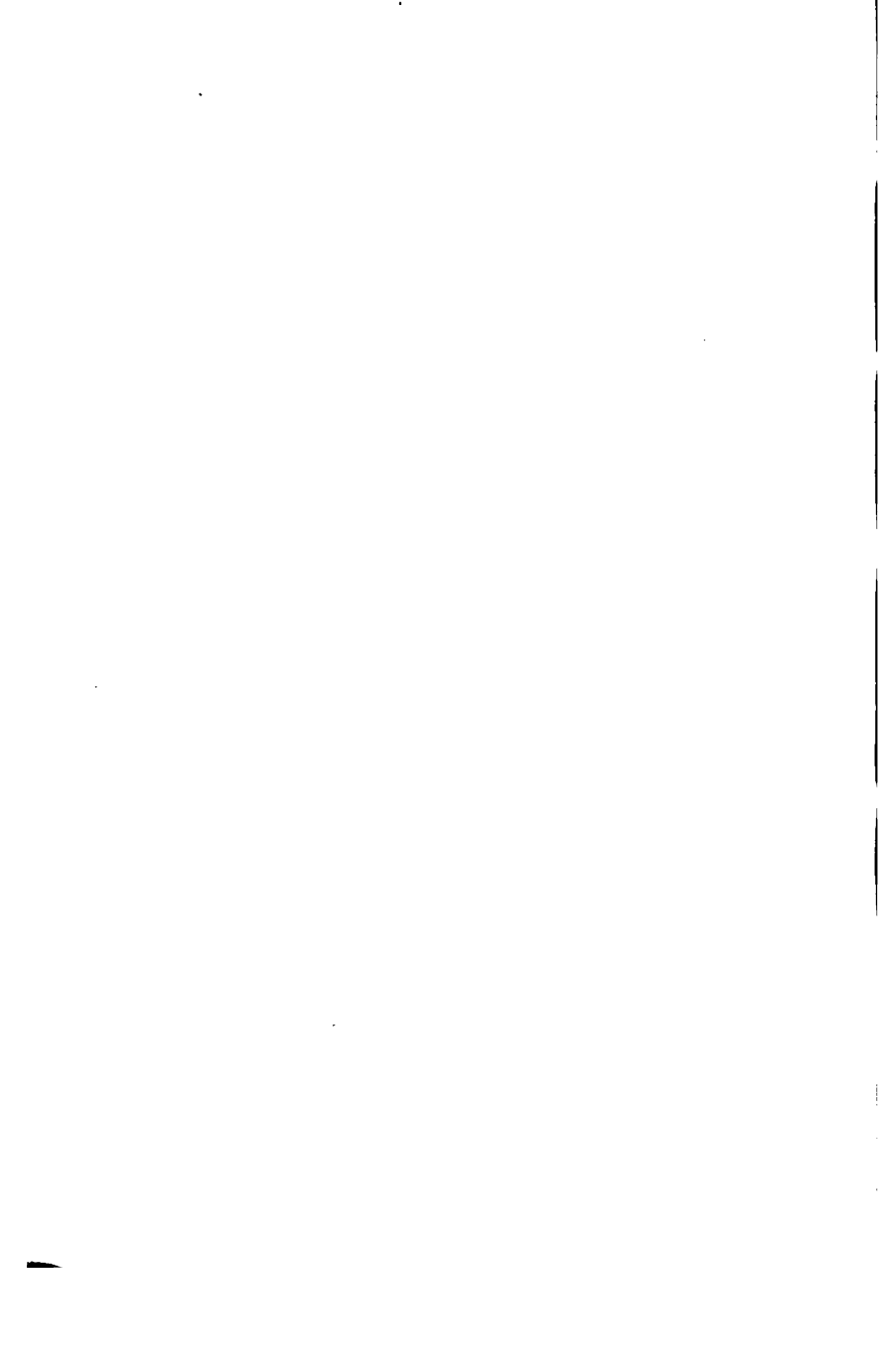
quantity of activity collected on a negatively charged rod was much more readily reduced by decreasing the pressure in small vessels than was the case when a vessel of greater diameter was used. This fact was brought out by the curves given in Fig. 25, in which curves I, II, and III give the results obtained with cylindrical vessels of diameters 1.1 cm., 2.6 cm., and 8.3 cm. respectively. The ordinates represent in arbitrary units the activity collected on the rod and the abscissæ the pressure in centimetres of mercury of the air with which the emanation was mixed.

The experiments referred to above prove without doubt that in the absence of air or any other gas the carriers of the active deposit are not attracted to the negative electrode. Looked at from the point of view of the kinetic theory of gases, we may say that a particle of active deposit must make a certain number of collisions with gas molecules before it can be directed by an electric field. At high pressures the particle makes sufficient collisions when it has travelled a very short distance, but at low pressures the



### PRESSURE IN CENTIMETRES OF MERCURY

Fig. 25.—Curves showing activity collected on negatively-charged rod immersed in radium emanation, when mixed with air at different pressures. Curves I, II, and III refer to cylindrical vessels of 1.1, 2.6, and 8.3 cm. diam. respectively.



particle has to travel a considerable distance before this is the case. This fact accounts for the curves obtained at low pressures with vessels of different sizes as has been demonstrated by Jackson,<sup>1</sup> who has developed this conception mathematically.

It may therefore be considered as beyond doubt that it is necessary for a particle of the active deposit to make a certain number of collisions with gas molecules before it is capable of being directed by an electric field. The part played by the gas remains, however, somewhat obscure, for it is at present impossible, from the available experimental evidence, to decide whether the active deposit actually acquires its charge by collision with the air molecules or whether the active deposit, which when formed from the emanation must be projected with considerable velocity, merely has its velocity reduced by collision to a value sufficiently small for it to be capable of being influenced by the electric field in which it is produced. The solution of this somewhat complicated problem must await further experiments.

<sup>1</sup> W. H. Jackson, *Phil. Mag.*, November, 1905.

CONCENTRATION OF ACTIVE DEPOSIT ON THE CATHODE  
IN FIELDS OF DIFFERENT INTENSITIES

Henning<sup>1</sup> has made a careful study of the conditions which affect the deposition of the active deposit from thorium emanation, and has shown that the quantity of deposit obtained on a rod exposed to the emanation in an electric field depends, *ceteris paribus*, on the voltage which is applied to the rod. As the rod is successively raised to increasing negative potentials, the quantity of deposit collected on it in a given time increases until a certain value is reached, when a further increase of potential produces no further effect. When the amount of active deposit collected from a given quantity of emanation is plotted against the strength of field to which it is exposed, the curve obtained resembles the curves representing the relation between the electromotive force and the current flowing between two electrodes immersed in a gas. As in the latter so in the former case the curve approaches a "satura-

<sup>1</sup> Henning, *Annalen der Physik*, VII, p. 562, 1902.



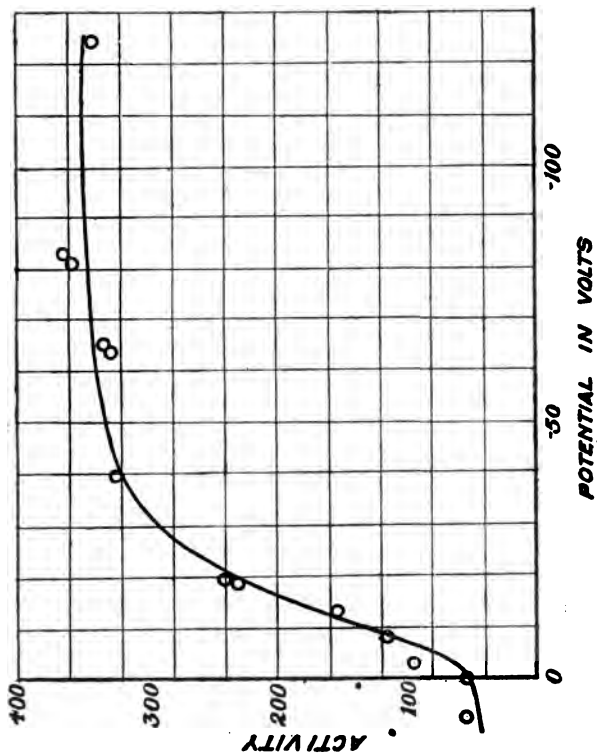


Fig. 26.—Variation with potential of amount of active deposit on charged wire in air at atmospheric pressure.

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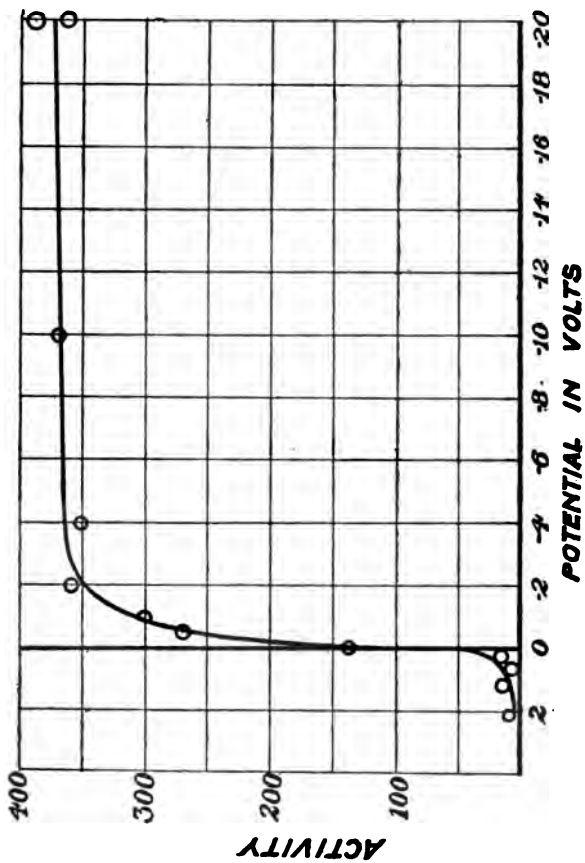
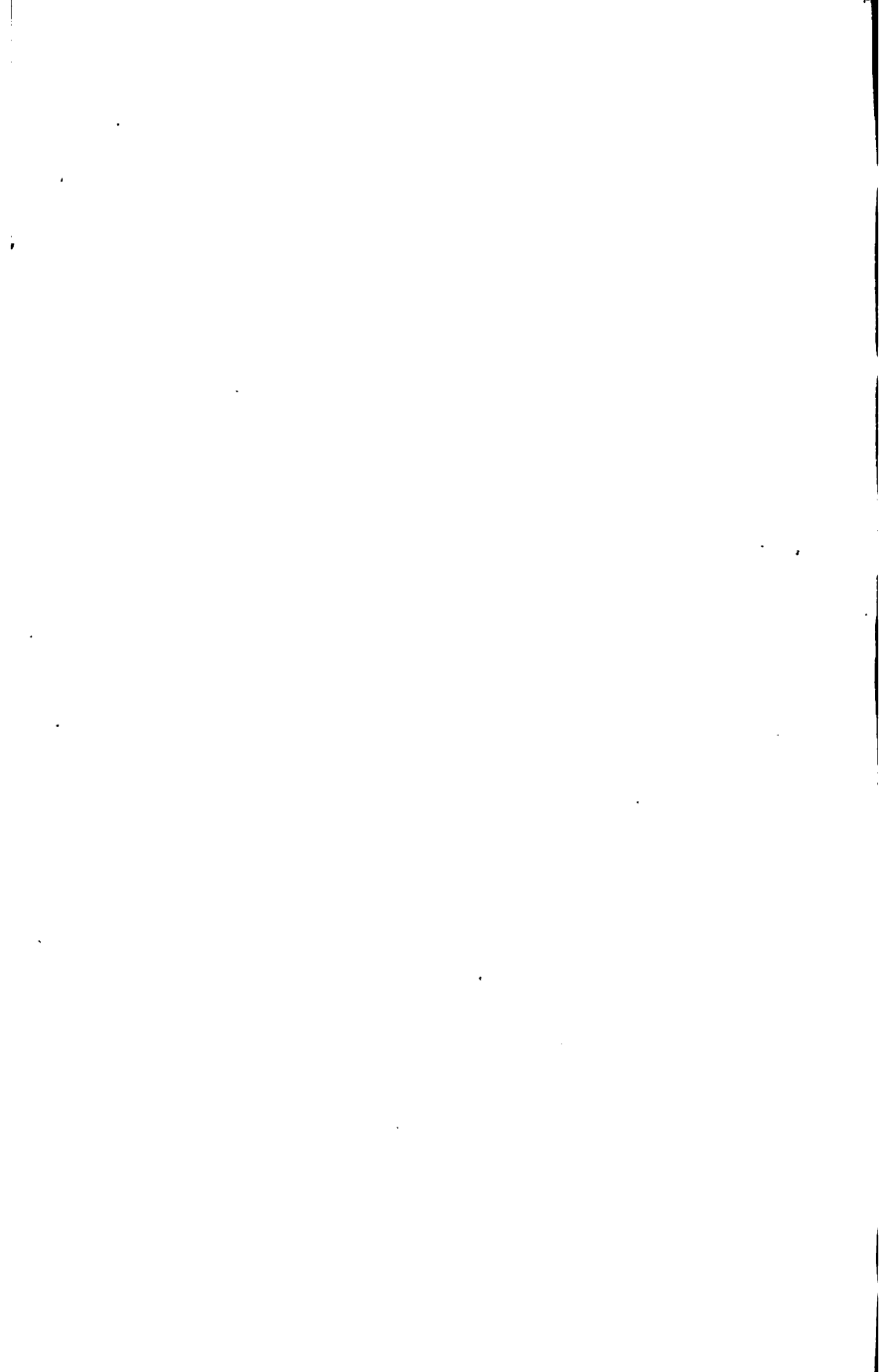


FIG. 27.—Variation with potential of amount of active deposit on charged wire in air at pressure of 1 cm. of mercury.



tion" value as the potential is increased. Similar results have more recently been obtained by the author with the deposit from radium emanation, and the observations somewhat extended by making experiments at different pressures. In Figs. 26 and 27 the results obtained at atmospheric pressure and 1 cm. mercury respectively are represented graphically. The general features of the curves are similar to those discovered by Henning. It will be noticed that at the lower pressure the curve reaches "saturation" much more rapidly than at the higher pressure. In the former case a negative potential of 2 or 3 volts sufficed to ensure "saturation," whereas in the latter case several hundred volts were required.

It has already been mentioned that in the case of radium a small portion of the activity travels to the positive electrode in an electric field, whereas this is not the case with thorium. Some recent experiments of Russ with radium have shown that whereas the amount of active deposit obtained on a negative electrode diminishes as the pressure is

decreased, the amount obtained on a positive electrode increases under the same conditions.

#### DECAY OF ACTIVE DEPOSIT WITH TIME

In dealing with the radioactive emanations we saw that the activity of these bodies, when removed from the radioactive bodies that are producing them, diminishes with time, slowly in the case of radium emanation and very much more rapidly with thorium and actinium emanations. Moreover, the activity of these bodies decayed according to an exponential law, the time taken for each emanation to lose half its activity being a constant quantity, by measuring which the emanation could be identified. The active deposits which we are studying, behave in a similar manner, losing their activity when separated from the emanations which produce them, and it would be unnecessary to do more than mention the fact, were it not that these bodies behave in a more complicated manner than the emanations.

In general it is found that the decay of

activity of the active deposits cannot be represented by a single exponential law, but that the curves of decay are not only more complicated but depend considerably on a variety of circumstances, such as the time during which the activity deposited from the emanation has been allowed to accumulate before testing and the particular rays by which the activity is measured.

Only by a very careful study and analysis of these decay curves has it been possible to interpret the results and throw new light on the constitution of the radioactive deposits. We will consider in some detail the investigations which have been carried out with radium in this direction, merely recording the conclusions which have been arrived at with thorium and actinium by similar experiments and reasoning. If, for example, a wire is exposed to radium emanation, we can study the rate of decay of the active deposit by removing the wire from the presence of the emanation and measuring the ionization produced in a gas at known intervals of time after withdrawing the wire

from the emanation. The time for which the wire is exposed to the emanation can be varied and the effect of this change on the results obtained may also be studied. Experiments of this kind were first made by Curie and Danne,<sup>1</sup> who discovered the main features of the phenomena involved. Miss Brooks<sup>2</sup> has investigated the subject in somewhat greater detail, and has succeeded in elucidating some of the obscurities attending the interpretation of the results given by Curie and Danne. The curves showing the rate of decay of the active deposit under different conditions are given in Figs. 28, 29, 30, and 31.

From Figs. 28-32 it will be at once seen that the form of the decay curves depends on at least two conditions. In the first place different curves are obtained when the time for which the surface subsequently to be tested is exposed to the emanation; and in the second place the characteristics of the curves are quite different if the measurements

<sup>1</sup> Curie and Danne, *C. R.*, CXXXVI, p. 364, 1903.

<sup>2</sup> Miss Brooks, *Phil. Mag.*, September, 1904.



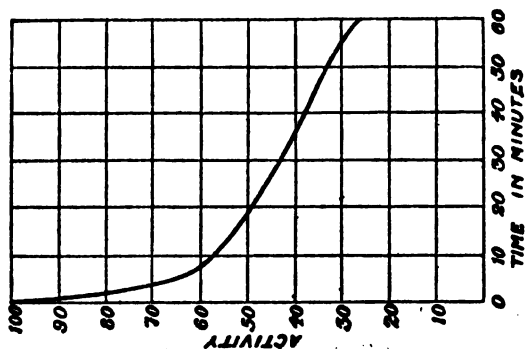


FIG. 28.—Decay of active deposit on plate exposed for long time to radium emanation, measured by Alpha rays.

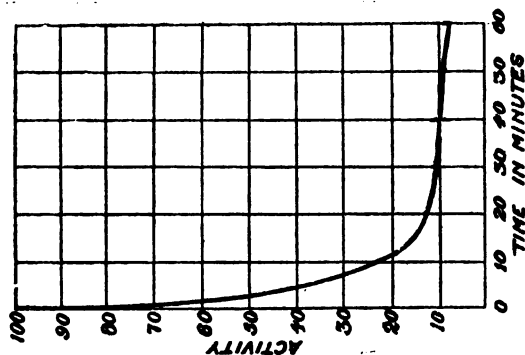
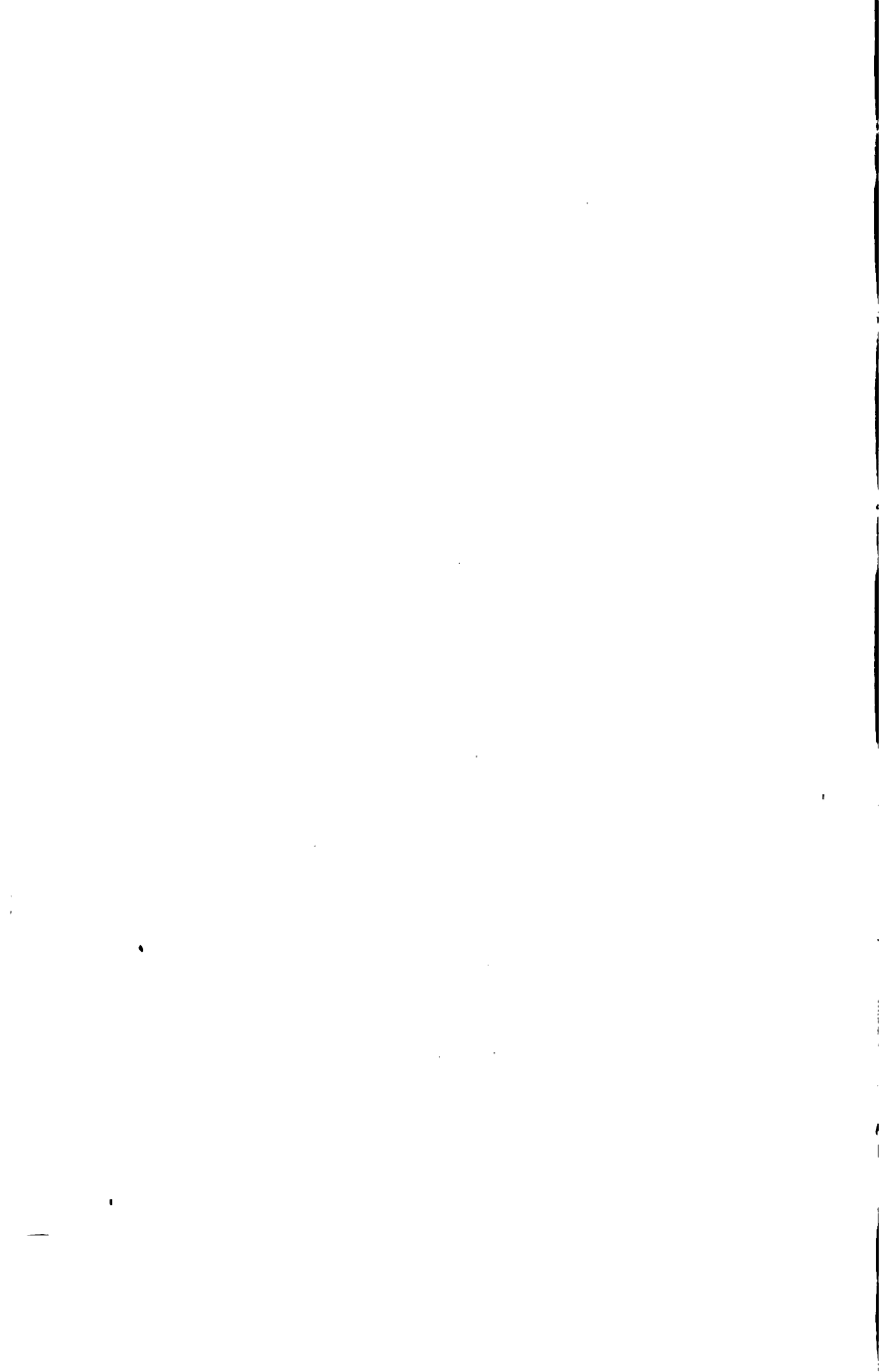


FIG. 29.—Decay of active deposit on plate exposed for short time to radium emanation, measured by Alpha rays.



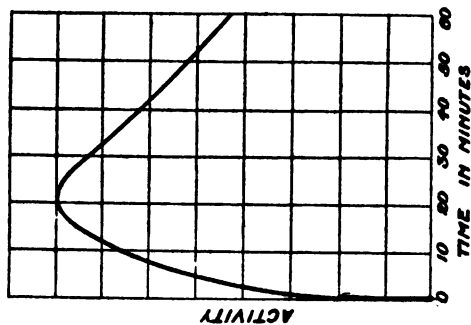


FIG. 30.—Decay of active deposit on plate exposed for short time to radium emanation, measured by Beta rays.

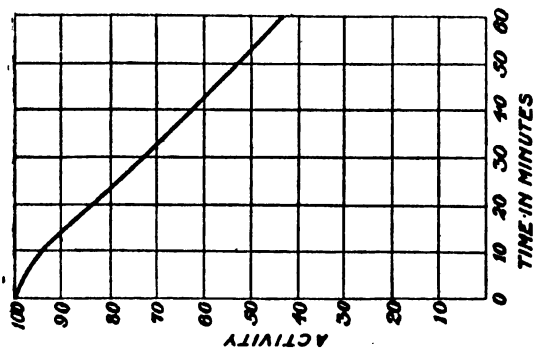
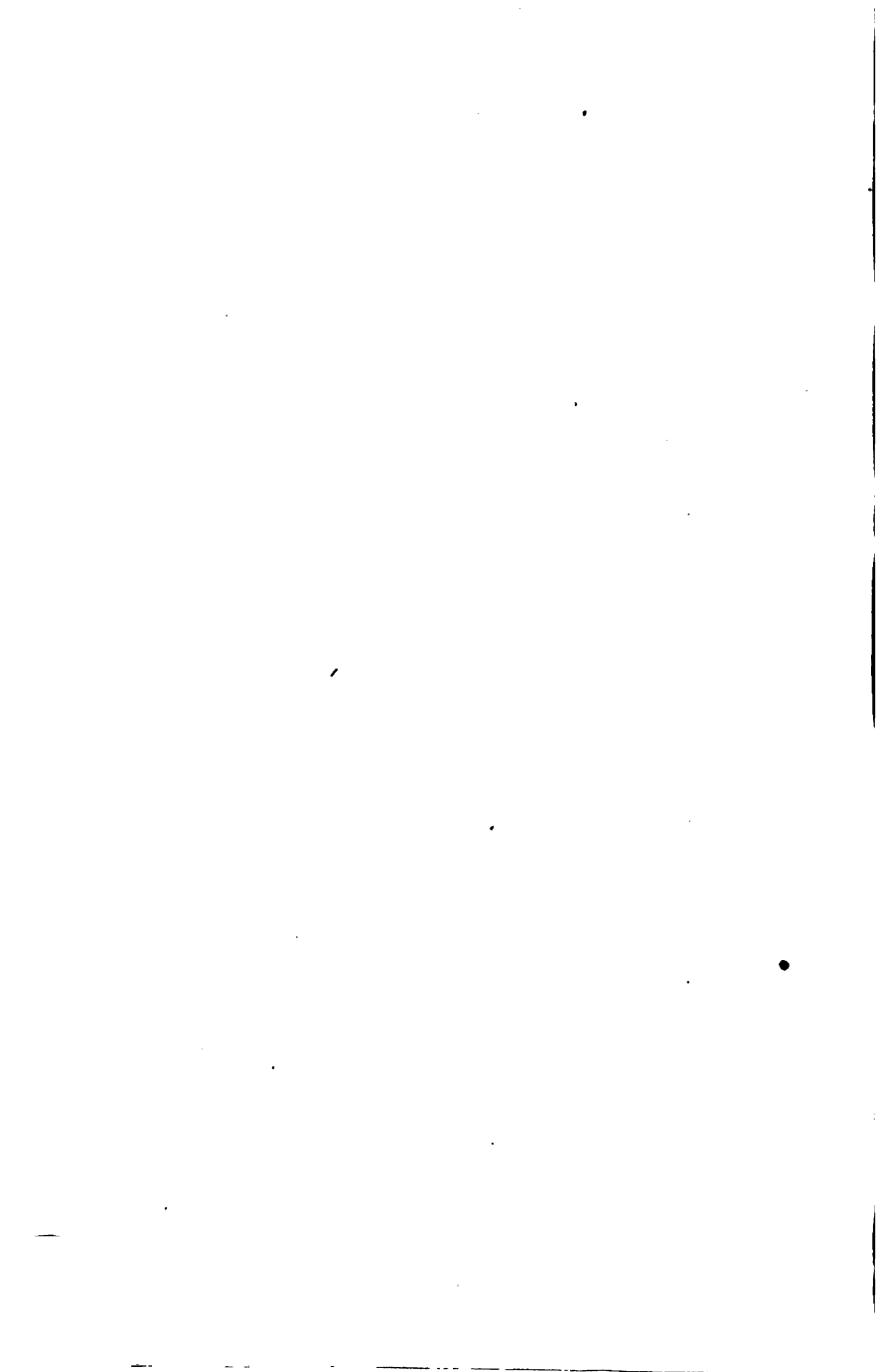


FIG. 31.—Decay of active deposit on plate exposed for long time to radium emanation, measured by Beta rays.



are made by means of the  $\beta$  or  $\gamma$  rays instead of by the  $\alpha$  rays.

Let us first consider the curves found from measurements with the  $\alpha$  rays. It will be noticed that when the active surface has been exposed to the emanation for only a short time the curve is very clearly divided into three portions (Fig. 29). During the first few minutes the activity falls off very rapidly with the time; this stage is followed by one during which the activity changes very little, if at all, after which the activity again begins to decrease though not as rapidly as during the first few minutes. As the time of exposure to the emanation is increased the three stages in the decay curves become less marked as is seen from Fig. 28. The fact that the  $\alpha$  ray curves after a short time of exposure to the emanation show these three very characteristic stages has given rise to the theory that the phenomena under investigation consist not of *one* radioactive process as in the case with the emanation, but of *three* such processes following in rapid succession upon each other; in other words, that we are actu-

ally dealing with three consecutive radioactive transformations. In order to follow the con-

sequences of this theory we will make use of an analogy in which the conceptions involved are more familiar.

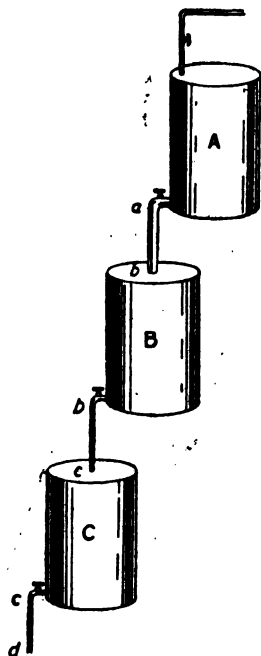


FIG. 32. — Hydrodynamic analogy to explain decay curves of active deposit from radium.

Suppose that we have three reservoirs, A, B, C, one above the other as shown in Fig. 32, into the uppermost of which water can be poured. Next let the reservoirs be connected by tubes *ab*, *bc* respectively, and let the reservoir C be provided with an outflow tube *cd*. Suppose further that the pipes *ab*, *bc*, *cd* are of different diameters and lengths, and that

things are so arranged that the resistance to the flow of water through *ab* is considerably smaller than the resistance to the flow

through cd, which is itself somewhat smaller than the resistance to the flow through bc. If the cross section of each reservoir is the same, then the quantity of water flowing through each of the tubes ab, bc, cd will be proportional to the quantity of water in the reservoir immediately above it. In order to make our analogy correspond more nearly with the case of the three radioactive changes, we will suppose that we have no means of determining the quantity of water in each reservoir, but that we have some measuring instruments, say water-meters, by means of which we can measure the *sum* of the currents of water flowing through ab and cd at any time. Now let water be poured into A at a constant rate for a known time, after which it is cut off. If the water has been poured into A for only a *short time*, there will be a large rush of water through ab for a short time after cutting off the supply flowing into A. There will, however, be very little water flowing through cd at first, because some time will elapse before an appreciable quantity of water has accumulated in B and run into C

to produce the current in cd. At first, then, the sum of the currents flowing through ab and cd will be large and will rapidly fall off, for, be it remembered, the tube ab is supposed to be wide compared with either bc or cd. Our water-meters will therefore show a rapid decrease in the current which they are measuring. During this time, however, water will be accumulating in the reservoir C, and consequently the current flowing through cd will be increasing. Hence, since we can, by hypothesis, measure only the sum of the currents of water passing through ab and cd, a time will ultimately be reached when the increase in the current through cd will exactly neutralize the decrease in the current flowing through ab, and our water-meters will for a short time indicate a constant current of water flowing through the two pipes. If we now wait still longer, the current passing through cd will itself begin to diminish when the rate at which water is being supplied to C is less than the rate at which it is flowing out through cd. Now since the current of water through ab is always decreasing, it follows



that the sum of the quantity of water flowing through *ab* and *cd* will again begin to diminish, at first slowly and then more rapidly.

The case is somewhat different if water is allowed to flow into the reservoir *A* *for a long time* before disconnecting it from the source of supply, for now the reservoirs *B* and *C* will become partially filled before disconnecting *A* from the source of supply. Consequently the current through *cd* will be comparable with the flow through *ab*, and although the sum of the currents through *ab* and *cd* will at first diminish more rapidly than later, on account of the rapid diminution of the part contributed by the current through *ab*, yet the stage will be much less marked than in the case previously considered. Moreover, the stage for which the sum of the currents through *ab* and *cd* is constant will no longer be so marked, since the level of the water in *C* in the present case rises but little after disconnecting the reservoir *A* from the source of supply.

Let us now apply the above reasoning to

the case of three radioactive products produced successively from each other as measured by the  $\alpha$  radiation emitted by them. The radiation emitted by each product is proportional to the quantity present at any moment, and therefore corresponds in our analogy to the current of water passing through the three pipes ab, bc, cd. If it so happened that the first and last products alone emitted  $\alpha$  rays and that the rate of decay of the first product was rapid compared with that of the third, which itself was slightly greater than that of the second, then we should have a state of affairs which exactly corresponded with the case of the three reservoirs considered. If an electrometer is employed to measure the total radiations given out by the three bodies, it measures the sum of the radiation emitted by the first and third body at any instant, since by hypothesis the second body is supposed to give out no  $\alpha$  radiation. The electrometer corresponds to the water current meter of our analogy, and the readings of the electrometer should show variations similar to those recorded by the water meter.

Thus suppose that we had exposed some body to the emanation for a very short time, the curve of decay, as measured by the  $\alpha$  rays, should at first show a rapid rate of decay which should be followed by a period during which the radiation remains nearly constant, after which the radiation would decay at first slowly and gradually more rapidly. Fig. 29, which shows the rate of decay of the active deposit, exhibits just these three characteristic portions. As in the case of the water analogy, if the body is exposed to a constant source of emanation for a long time the three stages in the decay curves will be less marked, and this is found to be the case (Fig. 28).

Let us now turn our attention to the case of the decay curves obtained by measuring the activity of the deposit by means of the  $\beta$  and  $\gamma$  rays, and let us assume that it is the third product which alone emits these rays. This case would correspond in our analogy to the case in which we were measuring the variation of the current of water through ed alone. It is obvious that in this case, if water is allowed to flow into A for a short

time only, then since the supply is cut off before an appreciable quantity of water has had time to accumulate in the reservoir C, the water meter will at first register hardly any flow of water; but this current will gradually increase until after some time the quantity of water entering the reservoir C will be less than that leaving it; the level of the water in C will then begin to diminish and a smaller current of water will flow through cd. If on the other hand the water is allowed to flow into A for a long time before disconnecting the supply, the level in C will no longer rise after disconnecting the supply, and the current through cd will at once diminish, at first slowly and then more rapidly. These features are manifested by the  $\beta$  and  $\gamma$  ray curves (Figs. 30 and 31).

From the analogy just considered we learn that the characteristics of the decay curves of the active deposit produced by radium emanation can be explained if we assume that the former consists of three radioactive products successively changing into each other, the first of which emits  $\alpha$  rays only, the second no rays

at all, and the third  $\alpha$ ,  $\beta$ , and  $\gamma$  rays (for the curves obtained when measurements are carried out with the  $\gamma$  rays are exactly similar to those shown with the  $\beta$  rays). The view that this is the real explanation of the observed phenomena was put forward by Rutherford, and we shall see later that the indirect reasoning from which this hypothesis has arisen is amply confirmed by direct experiment. The three products have been named respectively, radium A, radium B, and radium C.

Perhaps the most remarkable thing in connection with this view is the fact that it is necessary to assume the existence of a substance radium B which possesses all the properties of a radioactive body without itself emitting any radiation. This implies that if it were possible to isolate this particular product we should be unaware of its presence, for it will be remembered that it would be impossible to obtain a sufficient quantity of such a product for us to detect its presence otherwise than by the radiation which it emits. This difficulty has, however, been removed by the work of H. W.

Schmidt,<sup>1</sup> who has shown beyond the possibility of doubt that this product emits a kind of radiation, probably of the  $\beta$  type, slightly more penetrating than the  $\alpha$  rays, but much less penetrating than the  $\beta$  radiation emitted by radium C. In the previous experiments on which the theory is based, the conditions were such as to preclude the possibility of detecting these rays. The experiments of Schmidt therefore remove the chief difficulty in connection with the theory.

The reasoning which we have just considered serves to illustrate how the decay curves of the active deposit from radium may be deduced by considering the deposit as made up of three components. It is possible, however, to investigate the matter mathematically somewhat more precisely. If this be done, it can be shown that the decay curves obtained are the result of the combination of the three quantities varying according to the simple geometric law discussed in the chapter on the emanation, and that the curves of decay may

<sup>1</sup> H. W. Schmidt, *Physikalische Zeitschrift*, VI, p. 897, and *Annalen der Physik*, XXI, p. 609.

be obtained by assigning suitable values to the constants ( $\lambda$ ) referring to each change. The mathematical analysis, however, though comparatively simple, is beyond the scope of the present work. We can here, therefore, only record the result that it has been found that radium A decays to half its activity in about three minutes, radium B in about twenty-six minutes, and radium C in about nineteen minutes.

#### SEPARATION OF THE PRODUCTS RADIUM

##### A, B, AND C

The evidence which we have hitherto considered as to the complex nature of the active deposit from radium depends upon the analysis of the decay curves, and is of an entirely indirect nature. Confirmation of a more direct nature in support of the theory is, however, not wanting, for it is possible by suitable means to separate the three products, A, B, and C, from each other. This may be done by subjecting the active deposits to high temperatures when, on account of

the difference of volatility, the deposit may at any rate be partially separated into its constituents.

Miss Gates<sup>1</sup> was the first to show that the activity imparted to a wire might be removed by heating, and it was not long before Curie and Danne<sup>2</sup> extended these observations by showing that radium B was very much more volatile than radium C. In the experiments of the latter, radium B was completely removed from an active wire at a temperature a little over 600° C., whereas the radium C was not completely removed even at 1300° C. A fact, however, of even greater importance was brought out in these investigations, namely, that when an active wire was subjected to different temperatures between 600° C. and 1300° C. the rate of decay of the matter remaining on the wire depended on the temperature to which the wire had been heated. This is shown by the accompanying table given by Curie and Danne.

<sup>1</sup> Miss Gates, *Physical Review*, May, 1903.

<sup>2</sup> Curie and Danne, *Comptes Rendus*, CXXXVIII, p. 748, 1904.



TABLE I

Temperature centigrade			Time for activity to fall to half value
630°	...	...	29·3
830°	...	...	24·6
1000°	...	...	21·0
1100°	...	...	20·3
1250°	...	...	24·1
1300°	...	...	25·4

It will be seen that the rate of decay increases up to a temperature of 1100° C., reaches a maximum, and then decreases. The change was attributed to a real change in the rate of decay of radium C, and the result is of the utmost importance, since it is the only case known in which the rate of decay of any radioactive product has been altered by any change of physical or chemical conditions. This subject has already been alluded to.

Such, then, is the importance of the conclusions arrived at by Curie and Danne that it is not surprising that experiments have since been performed by other investigators to test the accuracy of these results. If the changes recorded in Table I are really due to an alteration in the rate of decay of

radium C, then the rate of decay of an active wire, sealed in a glass tube, should be affected by exposing it to a high temperature, although no volatile product was allowed to escape from the tube. A very careful series of experiments extending to a temperature of  $1100^{\circ}\text{C}$ . carried out by Bronson<sup>1</sup> failed to reveal any such change, and, therefore, threw doubt upon the conclusions reached by Curie and Danne. In view of these discrepancies a further investigation was undertaken by the author,<sup>2</sup> in the hope of throwing light on the matter. Experiments were made, not with the active deposit itself but with the emanation, for it was thought that if there were any change in the active deposit this could be detected by studying the radiation proceeding from a sealed tube, containing the emanation. Some radium emanation was therefore sealed in a quartz tube which could be heated to a high temperature in an electric furnace. After heating for a short time, the quartz tube was removed from the furnace and

<sup>1</sup> H. L. Bronson, *Phil. Mag.*, January, 1906.

<sup>2</sup> W. Makower, *Proc. Roy. Soc., A*, Vol. LXXVII, 1906.

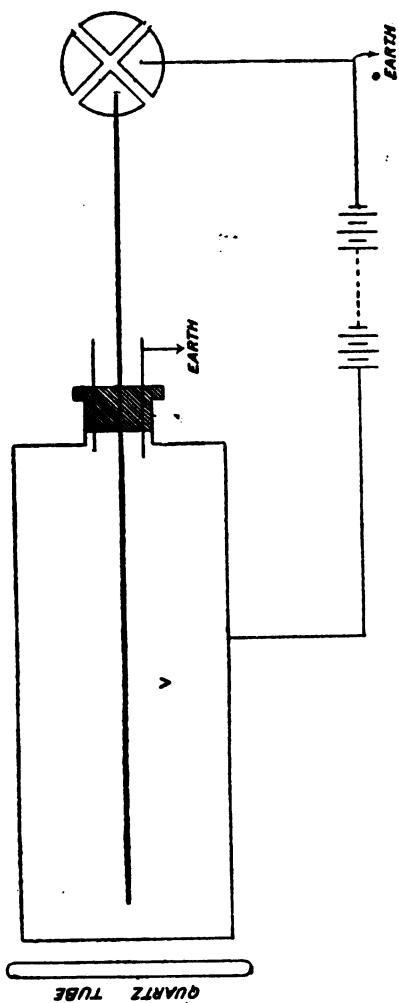
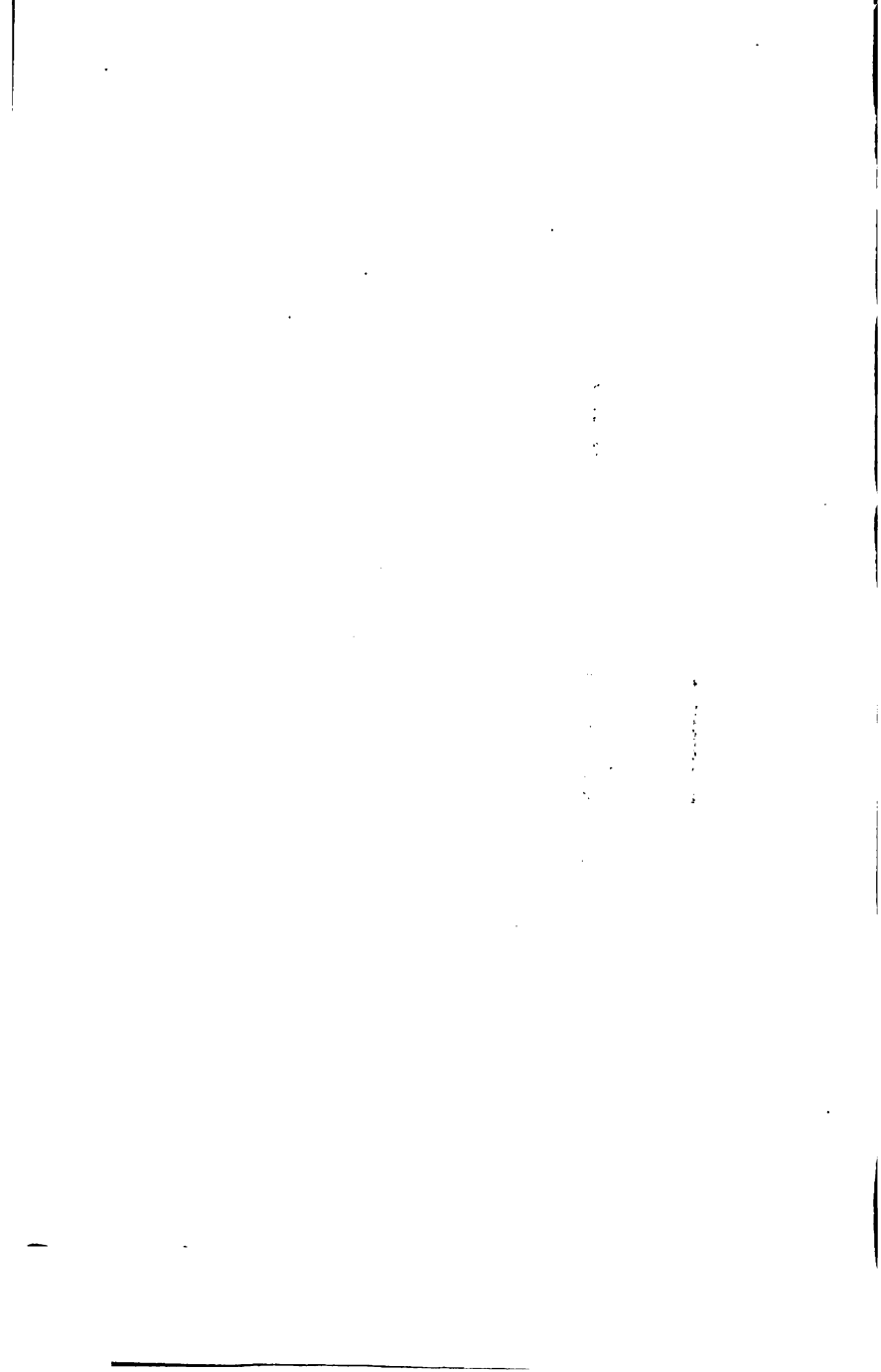


FIG. 83.—Author's experiment on change of activity with high temperatures.



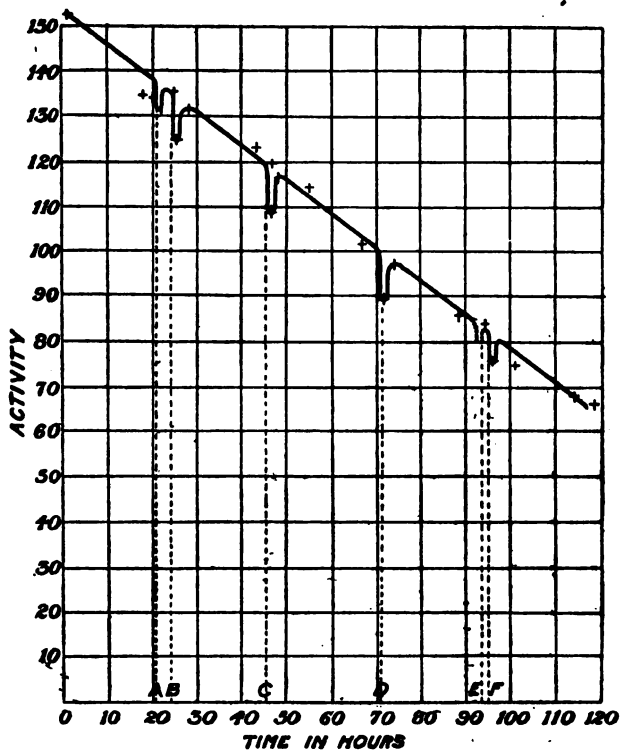
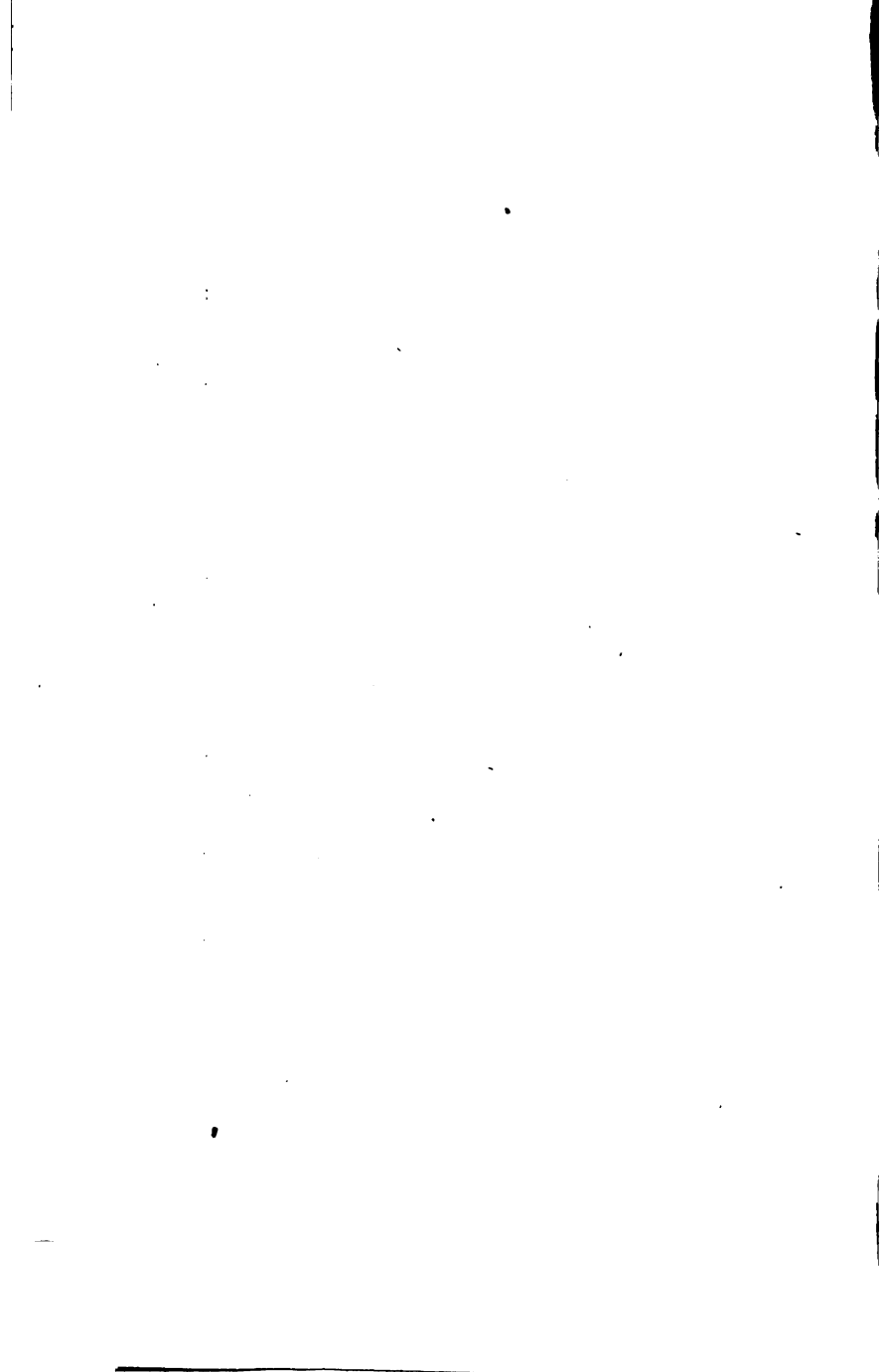


FIG. 34.—Diagram of effect of high temperature on radium emanation.



its activity tested by the ionization which it produced. The arrangement of the experiment is shown in Fig. 33. As a result of these experiments it was found that the activity always fell somewhat after the tube had been heated, but it was found that the activity gradually regained its normal activity within an hour or two of removing it from the furnace. The results are shown in the accompanying diagram (Fig. 34), the points A, B, C, D, E, F indicating the times at which the tube was heated. It will be seen that heating the tube always produces a temporary fall in the activity superposed on the normal decay curve of the emanation.

These results have since been criticized by Bronson,<sup>1</sup> who attributes the change observed to the fact that the quartz tube was always removed from the furnace before testing its activity. A very careful series of experiments by this physicist revealed no detectable change in the activity when the activity of radium was tested without removing it from the

<sup>1</sup> H. L. Bronson, *Proc. Roy. Soc., A.*, Vol. LXXVIII, p. 494.

furnace even when heated to 1600° C. It seems, however, quite probable that the effect under discussion might be detectable only after removal of the active deposit, so that these experiments do not necessarily prove the absence of any change of activity. A further series of experiments by Russ and the author<sup>1</sup> on the effect of high temperatures on the active deposit have afforded further evidence of a change in the activity of radium C, first observed by Curie and Danne, and the results of these experiments are given. The

TABLE

Temp. of furnace		Percentage fall in activity		Temp. of furnace		Percentage fall in activity
°C.	...	...		°C.	...	
700	...	0	...	1020	...	5·2
885	...	3	...	1080	...	8·7
900	...	3	...	1080	...	15·6
935	...	11	...	1110	...	7·9
955	...	-3	...	1170	...	15·4
965	...	3	...	1185	...	14·2
1000	...	8·5	...	1250	...	8·2

available evidence would therefore, on the whole, indicate that the activity of radium C

<sup>1</sup> Makower and Russ, *Proc. Roy. Soc., A*, Vol. LXXIX, p. 158. 1907.



can be changed by subjecting it to a sufficiently high temperature. For a further discussion of the matter, however, the reader must be referred to the original paper quoted above.

#### SLOWLY DECAYING PRODUCTS OF RADIUM

It has been shown that radium, as a result of its radio-activity, gives rise to an emanation which in turn produces the active deposit which we have seen consists of three separate radioactive products which have been named radium A, B, and C respectively. We have yet to inquire what becomes of this active deposit when it loses its radioactive properties.

The first step towards the solution of this problem was made by Madame Curie,<sup>1</sup> who showed that, although the major part of the activity of the active deposit had disappeared within a few hours after removal from the emanation from which it had been produced, a small residual activity always remained which did not appear to decay with time.

<sup>1</sup> Mme. Curie, *Thèse*. Paris, 1903.

The activity left in this way was exceedingly minute, and did not amount to more than  $\frac{1}{20000}$  of the activity originally deposited, and on that account was not further investigated.

A recent investigation of this residual activity by Rutherford<sup>1</sup> has, however, thrown light on the matter, and has resulted in the discovery of yet three more radioactive bodies. The experiments and reasoning which have led to the discovery of these products are analogous to those from which the existence of radium A, B, and C was inferred, and it will, therefore, suffice to briefly indicate the way in which the existence of these three new bodies has been proved and the methods by which their properties have been investigated.

It was found in the first place that if the active deposit collected on a platinum plate by a long exposure to a large quantity of radium emanation was left for a sufficient time for radium C to completely vanish, the residual activity as measured by the  $\alpha$  rays

<sup>1</sup> Rutherford, *Phil. Mag.*; November, 1904.

very gradually increased for a period of eighteen months, after which time, although it was still increasing, the activity appeared to be approaching a limit. On the other hand, if the activity was measured by the  $\beta$  rays which were emitted instead of the  $\alpha$  rays, the rise of activity was very much more rapid, rising to half its final value in about six days, and reaching a sensible maximum after about forty days. This difference of behaviour of the activity when measured by the  $\alpha$  and  $\beta$  rays is very striking, and at once gave rise to the supposition that these two types of radiation are emitted by different products produced by some parent substance which, like radium B itself, gives out little or no detectable radiations. If this supposition is correct, it is evident that this parent substance must itself decay at an exceedingly slow rate. Just as in the case of radium A, B, and C, the indirect evidence deduced from an analysis of the decay curves is supported by direct experiment. It was found that, by subjecting the residual activity under consideration to high temperatures, the  $\alpha$  ray and  $\beta$  ray ac-

tivity could be almost completely separated, and the rates of decay separately studied. A temperature of  $800^{\circ}\text{C}$ . had little or no effect on the composition of the activity, but a temperature a little above  $1000^{\circ}\text{C}$ . almost completely removed the  $\alpha$  ray activity. At still higher temperatures the substance which gave rise to the  $\beta$  radiation appeared also to be slightly volatile. It was further shown that the  $\alpha$  ray activity could be separated from a solution of the active matter in dilute sulphuric acid. A study of the rates of decay of the  $\alpha$  and  $\beta$  ray activities when separated revealed that the former decayed to half value in six days.

From these experiments Rutherford concludes that the activity consists of three products, the first of which gives out no radiation, and decays with an exceedingly slow period, being half transformed in about forty years; the second emits  $\beta$  rays, and is half transformed in six days; while the third emits only  $\alpha$  rays, and is half transformed in one hundred and forty-three days. The three substances are called respectively radium D,

E, and F, and are supposed to be formed in succession from radium C. The experiments of Rutherford just mentioned thus revealed the existence of three new products produced by radium, the time periods of which were also measured. But there was a difficulty in connection with the determination of the period of radium E which is worthy of mention.

If observations were made on the residual activity after radium C had lost its activity, it was found that the activity as measured by the  $\beta$  rays continued to increase for more than a month before attaining a steady value. This, as we have seen, is due to the formation of radium E from radium D, whose period of decay is very long. An examination of the curve giving the increase of the  $\beta$  ray curve with time showed that half of the final value of the radiation was attained in six days, from which it can be inferred that radium E, if separated, would lose half its activity in six days.

Now radium E can be separated from D, as was shown by Rutherford, by subjecting the mixture to a temperature of 1000° centigrade.

At this temperature radium D is volatilized and E is left behind, and the rate of decay of this latter body can now be directly investigated. When this was done it was found that the activity did not fall to half its original value in six days as had been expected, but decayed more rapidly, being reduced to half in 4.5 days. This was somewhat surprising, as the discrepancy was too great to be attributed to experimental errors.

The difficulty has since been removed by the work of Meyer and Schweidler,<sup>1</sup> who have shown that radium E consists of two products, the first of which gives out no rays and is half transformed in a little more than six days, and a second product which emits  $\beta$  rays and is half transformed in 4.8 days. These two products have been named by them radium E<sub>1</sub> and E<sub>2</sub>, respectively.

#### THE CONNECTION BETWEEN RADIUM F AND POLONIUM

It will be remembered that a substance was obtained by Marckwald from radioactive bis-

<sup>1</sup> Meyer and Schweidler, *Wien, Sitzungsab.*, CXV, 11a, 697, 1906.

muth, obtained from pitchblende by immersing a rod of bismuth in it (chap. III, p. 60). This substance was named radio-tellurium and has since been shown to be identical with polonium. Now we have just seen that radium F can be separated from a solution of the active deposit of slow decay by an exactly similar process. Moreover, the radium F so separated decays to half its activity in 143 days, a time which corresponds exceedingly closely with that of polonium. There would, therefore, appear to be little doubt that the newly-discovered radium F is nothing else than polonium. The discovery of this new radioactive product of radium shows the close connection which exists between the two well-known radioactive constituents of pitchblende, viz. radium and polonium.

## CHAPTER VIII

### THE HEAT EMITTED BY RADIOACTIVE BODIES

WE have studied at some length the properties of the radiations given out by radioactive bodies and have seen that it is possible to detect the radiations by the power that they possess of ionizing gases and by their photographic and fluorescent actions.

Now the power of producing these effects implies an emission of energy which must be supplied alike to ionize a gas, to affect a photographic plate, and to cause a screen of barium platinocyanide to become luminous. It is therefore necessary to consider whence this supply of energy is drawn and how it is maintained. When we remember that bodies such as radium are capable of emitting radiations of practically undiminished intensity for prolonged periods extending over years it will be realized that the question is one of fundamental importance.



The measurement of the energy emitted by radioactive bodies has been the subject of many investigations by different methods. We have seen that the energy of the radiations emitted by a body is used up in producing ionization in passing through a gas. Now if an estimate can be made of the energy necessary to produce an ion in a gas, then, by measuring the number of ions generated by radiation of known intensity, the energy emitted by a known quantity of a radioactive substance can be determined. The problem has been attacked from this point of view by Rutherford and McClung,<sup>1</sup> who deduced that one gramme of uranium oxide emitted energy at least at the rate of 0.032 gramme calories per year.

There is, however, a more direct method, which may be applied in the case of the more powerfully active bodies such as radium and its products. If, for example, a quantity of radium is contained in an enclosure from which the radiations are prevented from escaping, then the energy emitted by the

<sup>1</sup> Rutherford and McClung, *Phil. Trans.*, April 25, 1901.

radium remains within the enclosure, and is ultimately dissipated into heat. Consequently measurements of the heat developed by a radioactive body under such circumstances afford an estimate of the energy emitted. This plan has been adopted by several investigators, and results of the greatest importance have been obtained. At the beginning of the year 1903 Curie and Laborde<sup>1</sup> discovered that radium salts spontaneously develop sufficient heat to raise and maintain their temperature appreciably above that of surrounding objects. Thus, for instance, in one series of experiments the heat produced by a radium salt was compared with that generated by an electric current flowing in a coil of wire, and in another series the heat generated by radium was measured by means of the well-known ice calorimeter invented by Bunsen. Subsequent experiments have also been made by Runge and Precht,<sup>2</sup> and by Precht<sup>3</sup> alone, using the same

<sup>1</sup> Curie and Laborde, *Comptes Rendus*, CXXXVI, p. 673. 1903.

<sup>2</sup> Runge and Precht, *Sitzungsb. Berl. Akad.*, p. 783, 1903.

<sup>3</sup> Precht, *Ber. der Deutsch. Phys. Ges.*, II, p. 101, 1904.

method. Later still, experiments on the subject have been carried out by Ångström,<sup>1</sup> who compared the heating effect of a radium salt contained in a calorimeter made of various metals with the heat developed by a known electric current in a coil of manganin wire of known resistance. But by far the most important experiments have been made by Rutherford and Barnes,<sup>2</sup> who have made a very thorough investigation of the whole question, using as measuring instruments in some cases a differential air thermometer, and in others a platinum thermometer. The results obtained in the various researches quoted are given in the accompanying table:—

Authors		Heat emitted by 1 gramme of radium			
Curie and Laborde	. .	100	gramme calories	per hour	
Runge and Precht	. .	111	"	"	" "
Precht	. .	105	"	"	" "
Ångström	. .	117·8 to			
		114·3	"	"	" "
Rutherford and Barnes	. .	106 to			
		115	"	"	" "

<sup>1</sup> Ångström, *Arkiv. for Mat. Astr. och Fysik.*, I, p. 523 (1904) and II, p. 1 (1905).

Rutherford and Barnes, *Phil. Mag.*, Feb. 1904.

The results obtained by different experimenters are thus in good agreement, and the heating effect per gramme of radium may be considered as established with considerable accuracy. But Rutherford and Barnes have gone a step further and have determined the heating effect corresponding to each of the successive transformations taking place in radium.

For this purpose the emanation was separated from the radium by which it had been produced, and the heating effects of the emanation and radium measured separately. The heat produced by the radium was at first only slightly affected by the removal of the emanation; but whereas the heating effect of the radium fell off rapidly with time after removing the emanation, the heat produced by the latter increased correspondingly. This result could be explained by supposing that part of the heat produced by radium in equilibrium with its products was due to the active deposit. Hence when the emanation was withdrawn from the radium the first effect noticed was that the heat evolved by the radium

was reduced by an amount corresponding to that produced by the emanation. But on waiting a short time the active deposit left with the radium decayed, and since there was now no emanation to replace it, the heating effect fell to that corresponding to the radium itself. This amounted to about twenty-five per cent of the total heating effect of the radium when in radioactive equilibrium. It is easy to see that the heat developed by the emanation after separation from the radium should rise for a short time, and this was found to be the case. From their experiments Rutherford and Barnes were able to show that twenty-five per cent of the heat produced by radium in radioactive equilibrium was due to the radium itself, forty-one per cent to the emanation and radium A together, and thirty-four per cent to radium B and C. These numbers are approximately proportional to the  $\alpha$  radiations emitted by the several products, so that it may be concluded that the heat produced by radium is mainly due to the emission of  $\alpha$  particles.

PRODUCTION OF HEAT BY RADIUM IN THE SUN  
AND EARTH

The problem as to the origin of the heat radiated from the sun and the manner in which the temperature of the sun is maintained has long claimed the attention of men of science. For it may be shown that, in order to supply an amount of energy equal to that which is received by the earth from the sun, it would be necessary to burn about one ton of the best coal per hour per square foot of the sun. It is impossible to imagine that combustion is proceeding at this rate on the sun, and it is therefore clear that chemical action alone will not suffice to account for the heat which is constantly being emitted by the sun.

To overcome this difficulty the ingenious theory was suggested by von Helmholtz that the heat radiated might be supplied by the continual contraction of the sun. If the sun's radius contracted by one hundred and twenty-five feet per year, it was shown that sufficient energy would be liberated to account for all the heat known to be emitted by it. Thus this inappreciable contraction would

suffice to generate the heat necessary to maintain the sun at its present temperature.

The discovery of radium and the estimation of the heat evolved by this substance has opened up a new possibility with regard to the source of the energy of the sun. For it has been calculated by Rutherford<sup>1</sup> that if there are present in the sun 2.5 parts by mass in a million, this quantity would be sufficient to maintain the temperature of the sun in spite of the rapid dissipation of heat going on from the surface. There are certain difficulties in the way of accepting this hypothesis, for the presence of radium in the sun has never yet been revealed by spectroscopic analysis. Moreover, there is no means of ascertaining whether radium at the temperature of the sun would continue to emit heat at the same rate as under normal conditions on the earth. But it may be regarded as probable that if this rate is changed it would be greater and not less at the enormously high temperature to which the radium, if present in the sun, must be subjected. In any case the hypothesis is highly suggestive.

<sup>1</sup> Rutherford, *Radio-Activity*, p. 492.

Just as the presence of radium may play some part in supplying the energy radiated from the sun, so this substance may account for the high temperature maintained at the centre of the earth. Whereas in the case of the sun it is difficult to estimate the part played by radium, it is possible in the case of the earth to put the hypothesis to experimental tests. It can be shown that if there is present in the earth  $1.52 \times 10^{-13}$  grammes of radium per cubic centimetre, this would supply all the heat lost by the surface of the earth. As might be expected, this quantity is far less than the amount of radium which we have seen it is necessary to postulate in order to account for the heat emitted by the sun. By employing some data given by Elster and Geitel of the amount of emanation which diffuses out of the ground, Rutherford was able to show that there is probably at least the above-mentioned quantity of radium present in the earth.

The matter was of such great interest and importance and the available data so unsuitable for the purposes of the calculation




that experiments have since been carried out by Strutt<sup>1</sup> to obtain a more reliable estimate of the quantity of radium contained in the earth's crust. With this end in view a large number of rocks were examined for their radium content. A solution of the rock was stored till the emanation from the radium contained in it had had time to accumulate; the emanation was then removed by boiling, and its amount estimated by comparing the ionization produced by it with that produced by a standard quantity of emanation. Proceeding in this way, Strutt showed that in the rocks examined there were quantities of radium present varying from  $25.5 \times 10^{-12}$  to  $1.84 \times 10^{-12}$  grammes per cubic centimetre. Thus in all cases there is more radium present in the rock than would be necessary to supply the continuous loss of heat by the earth. This fact, then, opens up a new difficulty, for not only does there appear to be enough radium present in the earth to supply the heat which is escaping, but there is at least ten times as much as is necessary for this purpose, assum-

<sup>1</sup> Strutt, *Proc. Roy. Soc., A.*, LXXVII, p. 472.

ing that the radium is uniformly distributed throughout the earth. To account for this anomaly it is supposed that this latter assumption is not the case, and it has been suggested that the radium is all confined to a superficial shell 45 miles in thickness, and that the matter inside the earth is entirely free from radium. It is interesting to note that it has been deduced by Milne<sup>1</sup> from considerations of the velocity of earthquakes that the interior of the earth is composed of homogeneous matter surrounded by a crust of a different nature about 30 miles thick; a conclusion which is in agreement with the deduction drawn by Strutt from a study of the distribution of radium in the earth.

<sup>1</sup> Milne, Bakerian Lecture, Royal Society, 1906.

  
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## CHAPTER IX

### THE RADIOACTIVE TRANSFORMATIONS OF URANIUM, THORIUM, AND ACTINIUM

#### URANIUM

It has been pointed out in a previous chapter that the bodies radium, thorium, and actinium give rise to emanations, but that repeated and most careful searches have failed to reveal any analogous product from uranium.

Now the emanations are radioactive gases, and it can hardly be regarded as more than a matter of chance that the three substances, radium, thorium, and actinium, should all give rise to products which happen to exist in the gaseous state at ordinary temperatures. It need therefore afford no real cause of surprise that such a product should not exist in the case of uranium. If, however, our views as to the nature of radio-activity are correct, we must regard uranium as being transformed

into some other form of matter, and it becomes necessary to inquire whether any evidence of such a transformation exists. The evidence on this point was supplied by Sir William Crookes<sup>1</sup> soon after the discovery of the phenomenon of radio-activity.

It is well known that uranium can be precipitated from a solution of a salt by the addition of ammonium carbonate; the precipitate is, however, dissolved when an excess of the reagent is added. But if the experiment is performed with sufficient care it is found that there is a small light brown residue which remains undissolved. This residue was collected by Crookes and found on examination to be highly radioactive—indeed, more so than an equal quantity of uranium itself.

Just as the activity of radium is temporarily diminished by the separation of the emanation, so the activity of the uranium from which the brown precipitate had been separated was diminished in activity. The activity of the uranium, however, gradually increased with time, whereas that of the

<sup>1</sup> Crooke's *Proc. Roy. Soc.*, LXVI, 409, 1900.

brown precipitate, which Crookes called Uranium X, gradually lost its activity.

The phenomenon will therefore be seen to be exactly analogous to the separation of radium emanation from radium, except that in the present case the product, uranium X, separated from the uranium, does not happen to be gaseous. The rate of decay of uranium X was measured and found to lose half its activity in twenty-two days. It gives out only  $\beta$  and  $\gamma$  rays, whereas the uranium from which it has been separated gives out only  $\alpha$  rays.

Direct experiments have up till the present failed to reveal with certainty the presence of any other product from uranium, but there is considerable indirect evidence pointing to the conclusion that uranium X ultimately gives rise to radium and the series of radioactive bodies which we have already studied. This most important matter will be more fully considered in a later chapter.

#### THORIUM AND ACTINIUM

The important discovery made both by Hahn and Blanc of a highly radioactive sub-

stance which has been called radio-thorium, exhibiting all the radioactive properties of thorium, has already been mentioned in chapter III, p. 50. Long before this discovery, however, it was shown by Rutherford and Soddy<sup>1</sup> that a body analogous to uranium X could be separated from thorium. The separation was effected by precipitating the thorium from a solution of a salt by means of ammonia. The thorium so precipitated was found to have lost much of its activity, whereas the solution when evaporated to dryness was found to be strongly radioactive. This active residue, which by analogy with uranium was called thorium X, emitted only  $\alpha$  rays, and was found to decay to half its activity in about four days. It had the property of giving off thorium emanation, whereas the freshly precipitated thorium had temporarily lost that power.

It appears, therefore, that unlike radium, thorium does not give rise directly to an emanation, but that the formation of this gas is preceded by two changes, namely, from

<sup>1</sup> Rutherford and Soddy, *Journal of the Chemical Society*, LXXXI, p. 837, 1902.

thorium to radio-thorium, which in turn yields thorium X, which is the direct parent of the emanation.

The emanation from thorium gives rise to an active deposit in many ways analogous to that formed by radium emanation.

The nature of the deposit has been carefully studied, and it has been shown that it consists of two distinct bodies which have received the names thorium A and thorium B respectively, and which are successively formed from the emanation. The phenomena seem, therefore, to be less complex than is the case with radium.

The radio-activity of actinium has been studied and analysed by methods analogous to those used for the other radioactive elements. Suffice it to mention that the radioactive transformations of this element are closely analogous to those of thorium.

The accompanying tables give a complete scheme of all the known products of uranium, thorium, radium, and actinium, together with the time periods of each, and the radiations emitted by them.

	Time to fall to half value.	Nature of rays.
Uranium . . .	?	$\alpha$
Uranium X . .	22 days	$\beta, \gamma$

	Time to fall to half value.	Nature of rays.
Thorium . . .	?	$\alpha$
Thorium X . .	4 days	$\alpha$
Mesothorium .	1 year	?
Radiothorium .	109 years	$\alpha$
Emanation . .	53 secs.	$\alpha$
Th. A . . .	11 hours	no rays
Th. B. . . .	55 mins.	$\alpha, \beta, \gamma$

	Time to fall to half value.	Nature of rays.
Radium . . .	1300 years	$\alpha$
Emanation . .	3.75 days	$\alpha$
Ra. A . . .	3 mins.	$\alpha$
Ra. B . . .	26 mins.	slow $\beta$ rays
Ra. C . . .	19 mins.	$\alpha, \beta$ , and $\gamma$
Ra. D . . .	40 years	no rays
Ra. E <sub>1</sub> . . .	6 days	no rays
Ra. E <sub>2</sub> . . .	4.8 days	$\beta$ rays
Ra. F . . .	143 days	$\alpha$

	Time to fall to half value.	Nature of rays.
Actinium . . .	?	no rays
Radio-actinium .	19.5 days	$\alpha$
Actinium X . .	10.2 days	$\alpha$ ( $\beta$ ?)
Emanation . . .	3.9 secs.	$\alpha$
Actinium A . .	36 mins.	no rays
Actinium B . .	2.15 mins.	$\alpha, \beta, \gamma$



## THE ORIGIN OF RADIUM AND THE ULTIMATE DIS-INTEGRATION PRODUCTS OF THE RADIOACTIVE ELEMENTS

We have seen that the radioactive elements uranium, radium, thorium, and actinium all give rise to a series of products, and that we may regard these as arising from the disintegration of the elements from which they are derived. There are, however, certain difficulties connected with this theory of disintegration as to the nature of the phenomena involved in the changes which we have been studying, which must receive explanation before the theory can be accepted as correct.

It was pointed out when dealing with the formation of the emanation that the quantity of this gas formed from radium was so small that it would be quite hopeless to detect any change in the weight of the radium from which it was formed in any experiment yet performed. The case is, however, different if we consider the processes going on in a radioactive mineral, for it is known from geological data that these minerals have existed in their present state for many thou-

sands of years. It is therefore evident that although no appreciable quantity of radium disappears during the formation of the emanation during the time of any experiment yet performed in the laboratory, we are not justified in assuming this to be the case when dealing with minerals containing radium in which radioactive processes must have been going on for thousands of years. Indeed the reverse would seem to be more likely.

#### THE AGE OF RADIUM

A simple calculation will show that the radium contained in a mineral such as pitchblende must long since have been converted into the emanation and its subsequent products *unless the radium was being supplied from some source.*

For consider the emanation obtained from 1 gramme of radium with which it is in radioactive equilibrium. As soon as separated this emanation will begin to disappear according to the law given by the equation

$$N = N_0 e^{-\lambda t}$$

where  $N$  is the quantity of emanation at any time,  $N_0$  is the quantity just after separation from the radium, and  $\lambda$  is the constant of the decay of the emanation. Now since the emanation is half transformed in 3·8 days, we can at once calculate the value of  $\lambda$  from the above equation to be  $\cdot 21 \times 10^{-6}$  (sec)<sup>-1</sup>. Now we know from the experiments of Ramsay and Soddy that 1 gramme of radium would give rise to about ·001 cc. of emanation. And since the density of this gas is 50 times that of hydrogen, the density of which is ·0009 grammes per cubic centimetre, we see that

$$\begin{aligned} N_0 &= \cdot 001 \times 50 \times \cdot 0009 \\ &= 4\cdot 5 \times 10^{-5} \text{ grammes.} \end{aligned}$$

Now the weight of the emanation transformed per second is

$$\begin{aligned} N_0 \lambda &= \cdot 21 \times 10^{-6} \times 4\cdot 5 \times 10^{-5} \\ &= \cdot 8 \times 10^{-11} \text{ grammes.} \end{aligned}$$

Since the atomic weights of radium and the emanation are not very different, the weight of emanation transformed per second is not very different from the weight of radium

transformed in the same time when equilibrium exists between these two bodies. Hence about  $10^{-11}$  grammes of radium are being transformed per second into emanation in a mineral containing 1 gramme of radium.

From this reasoning it follows that in one or two thousand years from now half the radium, say, in pitchblende will have disappeared. We have thus indirectly found an estimate of the life of radium from a knowledge of the rate of decay of the emanation and its rate of formation from radium. There is other quite independent reasoning from which the life of radium has been deduced, and the result of such reasoning is in surprisingly good agreement with that obtained from the line of argument just given.

Without going into further detail we may therefore with fair certainty assume that in some time less than two thousand years from now half the radium at present in existence will have been converted into the emanation and its derivatives.

If now we work backwards and assume that this transformation of radium has been going

on in radioactive minerals throughout geological ages, we are driven to the conclusion that these minerals would long since have ceased to contain an appreciable quantity of radium, unless either the radium in them had been formed suddenly within comparatively recent times, or the radium which is decomposing is constantly being supplied from some unknown source.

Obviously the former hypothesis is exceedingly unlikely, so that *a priori* it is most probable that radium is being very slowly formed in order to make up for the quantity which we know to be constantly disappearing.

If, then, we are to retain the disintegration theory of radioactive processes, we see that it necessarily follows that radium is constantly in process of formation as well as decomposition, and we are confronted with the question whence this supply of radium is obtained.

Hopeless as it may at first sight appear to throw any light on this difficult question, much important work has already been successfully carried out with a view to solving the problem. It is suggestive that radium is

found in considerable quantities only in such minerals as pitchblende, which contain large amounts of uranium. This fact was recognized some time ago by Rutherford and Soddy, who pointed out that uranium, which is itself radioactive, might prove to be the source from which radium is being supplied.<sup>1</sup>

Let us consider the consequences of this hypothesis that radium is being produced by the disintegration of uranium. First imagine a quantity of uranium known to contain no radium. It is clear that if the uranium is producing radium, this latter substance will gradually make its appearance in the uranium, and the quantity of radium will increase as time proceeds. But it must be remembered that the radium itself begins to disintegrate as soon as it has been formed. Consequently the amount of radium present in the uranium will continue to increase until a certain time, when sufficient radium having been formed, the rate at which it is disappearing will just counterbalance the rate at which it is being produced from uranium. Now it has already

<sup>1</sup> Rutherford and Soddy, *Phil. Mag.*, May, 1903.

been demonstrated that the "life" of radium is only a few thousand years, so that we should expect that after a few thousand years the uranium with which we started would contain the maximum possible quantity of radium, which would then be disappearing at a rate just equal to the rate at which it is being supplied from the uranium. The case is, in fact, exactly similar to that of the formation of the emanation from radium, which has already been considered at some length. It was seen that it was possible to obtain only small quantities of the emanation, because after a few days the rate at which the emanation is decomposing into the active deposit is equal to the rate at which it is being formed. Imagine this process to take place about fifty thousand times as slowly, and we have the case of the transformation of uranium into radium.

If the hypothesis under consideration is correct, it follows that the quantity of radium present in different samples of uranium which have been kept for several thousands of years should bear a constant ratio to the quantity

of uranium in the sample. As samples we have the uranium ores which are known to have existed in their present state for a sufficiently long time to admit of the application of this test. We have therefore, at any rate, the means of testing indirectly the validity of this view of the origin of radium. If uranium ores are analysed we should expect the ratio of the quantity of uranium to that of radium to be always constant.

It is unnecessary to insist on the importance of this conclusion. The question as to its correctness has been taken up experimentally by several investigators, amongst whom we may mention McCoy,<sup>1</sup> Boltwood,<sup>2</sup> and Strutt.<sup>3</sup> McCoy analysed a number of uranium minerals and also compared their total radio-activity, i.e. the activity due to the uranium, radium, and its disintegration products. He found that the total

<sup>1</sup> McCoy, *Ber des Deutsch. Ch. Gesell*, XXXVII, p. 2641, 1904.

<sup>2</sup> Boltwood, *Phil. Mag.*, April, 1905.

<sup>3</sup> Strutt, *Proc. Roy. Soc.*, March 2, 1905; *American Journ. Sc.*, October, 1905, and December, 1906; *Phys. Zeitschr*, 1907, p. 98.



activity of any fixed quantity of a mineral was roughly proportional to the amount of uranium found to be present by analysis. This result is, of course, in accordance with the hypothesis that the radium present in the ore has been formed from the uranium. There were, however, certain conspicuous exceptions to these results. For ores which contained appreciable quantities of *thorium* the radioactivity was always greater than would have been predicted from a knowledge of the uranium content. In such cases, of course, the activity due to the thorium must be added to the activity theoretically deduced from the analysis, and it could therefore hardly be expected that such minerals would conform to the law shown to hold good for minerals free from thorium. The work of McCoy afforded the first experimental confirmation of the view expressed by Rutherford and Soddy as to the origin of radium; a wider and more searching investigation of the question was, however, necessary before the hypothesis could be accepted with any degree of certainty. A most careful and extensive

series of observations has been carried out by Boltwood, the results of whose experiments leave little, if any, doubt as to the origin of radium.

In an early communication on the subject, Boltwood describes a new and improved method of analysing the uranium minerals and a method of obtaining an estimate of the radium content by measurement of the total quantity of emanation obtainable from a known quantity of an ore. Since most of the emanation formed in a solid body remains occluded, a measurement of the total quantity of emanation evolved on dissolving the mineral and boiling the solution is proportional to the radium present. Using these new and improved methods, Boltwood was able to show that the ratio of the quantity of radium to uranium present in the twenty-two minerals which he investigated was sensibly constant. In fact, the discrepancies in its results were always within the limits of the experimental errors which might be expected in so difficult an investigation.

It may thus be taken as proved that uranium

gradually produces radium. We have, however, as yet no knowledge whether radium is formed directly from uranium, or whether there is some intermediate substance which is formed during the process of the transformation. Let us consider this point somewhat more closely, for it is obviously a matter of no little importance in the theory of radioactive transformation.

#### THE INDIRECT FORMATION OF RADIUM FROM URANIUM

Since it has been established from the evidence obtained from an examination of minerals which have existed in their present state for thousands of years, that radium has actually been in process of formation from uranium since the formation of the mineral, it is of interest to consider whether it would be possible to obtain any *direct* information of the transformation by experiment. The question which presents itself is, whether or not it would be possible to obtain from a sample of uranium a detectable quantity of radium in an experiment extending over a reasonable time.

Now it would be obviously impossible to observe any increase in the quantity of radium itself present in the uranium ; but it might be possible, by measurements of the change in the quantity of emanation obtainable from a solution of a uranium salt after standing for some months, to infer that there had been a formation of radium in the solution.

The test is an exceedingly delicate one, but is not beyond the range of experiment. For it can be shown by calculation that in every gramme of uranium  $10^{-9}$  grammes are transformed per year. Hence in one kilogramme  $10^{-6}$  grammes of uranium should be converted into radium in the course of one year, and the ionization produced by the emanation from this quantity of radium should be quite easily detectable by the refined instruments now in use. The experiment has actually been tried independently by Soddy,<sup>1</sup> Whetham,<sup>2</sup> and Boltwood,<sup>3</sup> who find that although radium gradually makes its appearance in a solution of a uranium salt kept in a closed vessel free from the possibility

<sup>1</sup> Soddy, *Nature*, May 12, 1904.

<sup>2</sup> Whetham, *Nature*, May 5, 1904.

<sup>3</sup> Boltwood, *Phil. Mag.*, April, 1905.

of contagion by any foreign radioactive matter, yet the amount of radium so formed is far less than would be expected on the assumption that radium is being *directly* formed from uranium. The discrepancy can, however, be explained on the assumption that some intermediate product of a slow rate of transformation exists between uranium and radium. It has been suggested that actinium, which is always found associated with minerals containing uranium, constitutes the necessary link between these two radioactive elements, and the question has quite recently been put to the test by Boltwood.<sup>1</sup>

Having first devised a method of chemical separation by which all the actinium in a mineral could be removed, a quantity of carnotite ore containing about twenty per cent of uranium was subjected to this process by which the actinium was removed from it. The solution of actinium chloride so obtained was sealed up in a glass bulb and tested at successive intervals for the presence of radium by measurements of the radium emanation

<sup>1</sup> Boltwood, *Amer. Jour. Sc.*, December, 1906.

evolved from the solution. Two months after the beginning of the experiment it was found that the amount of radium emanation present corresponded to the presence of  $5.7 \times 10^{-9}$  grammes of radium. The bulb was then again sealed up and allowed to stand for 193 days, at the end of which time the test was repeated. The solution of actinium chloride now contained  $14.2 \times 10^{-9}$  grammes, so that during the interval of time which had elapsed between the two experiments  $8.5 \times 10^{-9}$  grammes of radium had been formed in the solution. From this it follows that  $1.6 \times 10^{-8}$  grammes of radium would have been formed if the experiment had been continued for a complete year.

Now it had previously been ascertained that the solution originally contained 200 grammes of uranium, which is known to be associated with  $7.6 \times 10^{-5}$  gramme of radium when in radioactive equilibrium. From these data it is very easy to calculate the time constant of radium, for we have the equation

$$1.6 \times 10^{-8} = 7.6 \times 10^{-5} (1 - e^{-\lambda t})$$

in which the time is expressed in years.

Solving this equation we see that

$$\lambda = \cdot 00021 \text{ (years)}^{-1}$$

Expressed in other words the period required for the decay of radium to half its activity is 3300 years, a time which is in fair accord with the value of the "life" of radium obtained from quite different considerations.

The results of Boltwood are thus in agreement with the view that radium is being produced from uranium, not *directly*, but that the uranium is first transformed into actinium, from which the radium is then formed. But it has since been shown by Rutherford<sup>1</sup> that radium is not produced directly from actinium, but from a substance<sup>2</sup> which is usually precipitated with actinium, and which can be separated from the latter by suitable means.

Recent experiments by Boltwood have confirmed this fact, and it would seem that radium has no direct radioactive connection with actinium. The only outstanding diffi-

<sup>1</sup> Rutherford, *Nature*, June 6th, 1907.

<sup>2</sup> This new substance has been called *Ionium*.

culty with regard to the theory of the formation of radium from uranium is thus removed, and another important fact with regard to radioactive transformations established.

#### CONNECTION BETWEEN THE RADIOACTIVE ELEMENTS

We are now in a position to enumerate and classify the relations which exist between the known radioactive elements. It will be remembered that soon after the discovery of the radioactive properties of uranium by Becquerel, minerals which contained that element were found also to contain the following radioactive elements: *polonium*, *radium* and *actinium*, and occasionally *thorium*. In addition to these, another radioactive substance was discovered by Marekwald, which he called *radio-tellurium*, but which we have seen was subsequently shown to be identical with *polonium*. Both of these have since been shown to be the disintegration product of radium discovered by Rutherford, and which he called radium F.



The recent work of Boltwood and others has demonstrated that radium is an indirect disintegration product of uranium.

Thus with two exceptions all the radioactive elements known have been shown to belong to one single series of transmutations the one into the other. The two remaining exceptions to this scheme of radioactive transformations are thorium and actinium. As yet it has been found impossible to establish a relation between these elements and any other member of the series of transformations. Whether or not there is any foundation for the suggestion which has been made that thorium is, in fact, the parent of uranium, and thus constitutes the first member of the long series of transformations we have been considering, must remain for the future to decide. The fact that the atomic weight of uranium is greater than that of thorium is, however, a grave objection to such an hypothesis, which can only be true if the accepted atomic weight of one of these elements should prove to be incorrect.

The relations existing between the known

radioactive elements other than thorium is tabulated below :—

TABLE

Uranium	
↓	
Uranium X	
↓	
Ionium.	
↓	
Radium	
↓	
Radium Emanation	
↓	
Radium A	} Active deposit of quick rate of decay
↓	
Radium B	
↓	
Radium C	
↓	
Radium D	} Active deposit of slow rate of decay
↓	
Radium E <sub>1</sub>	
↓	
Radium E <sub>2</sub>	
↓	
Radium F (Polonium) (Radio-tellurium)	

#### THE ULTIMATE PRODUCTS OF RADIOACTIVE TRANSFORMATIONS

The transformations associated with the radio-activity of uranium have been traced through a long series of products, the last of which we have seen to be polonium. Now this element is itself radioactive and must, on the disintegration theory, give rise to some

form of matter distinct from itself. No such *radioactive* product has, however, as yet been discovered; nor does it seem probable that any such product of appreciable activity exists. The question therefore arises as to whether at the end of our series of radioactive elements, we have not at last arrived at a final form of matter which is not radioactive, and if so, whether this matter is identical with any known chemical element. The subject has given rise to considerable discussion, and investigations on the question have of late not been wanting; indeed, important information on this point has already been accumulated. In this matter, as in the question of the origin of the radioactive elements, we owe much of our knowledge to Boltwood.

Just as when dealing with the origin of the radioactive elements deductions could be drawn from a knowledge of the composition of ores containing uranium, so in the present case important facts have come to light as the result of a careful study of the composition of radioactive minerals. It is pointed

out by Boltwood that by the aid of geology and mineralogy it is often possible to assign the origin of a mineral to a definite geological period and therefore to form an estimate of the relative ages of various minerals. Now since it is known that the time which must have elapsed since the formation of the radioactive ores is exceedingly long, amounting probably to many million years, it is not unreasonable to expect that the final disintegration product of uranium should be found in them in sufficient quantity to be detected by ordinary chemical methods. Moreover, when analyses have been performed, it should be found that in *minerals of the same age* the ratio of the quantity of this final disintegration product to the quantity of uranium should be constant. Furthermore, the older the mineral the greater should be the relative proportion of the disintegration product we are looking for. Great caution is, however, necessary in the application of this method of investigation; for the validity of the reasoning used to interpret the results depends on the assumption that the mineral

in question *has undergone no change since the time of its formation*, a condition which in many cases is certainly not fulfilled.

It is usual to classify as primary those minerals for which the time of formation appears from available data to be synchronous with that of the formation of the material of the geological formation in which it occurs. On the other hand, those minerals which have been deposited at a later date through the action of percolating water or otherwise are spoken of as *secondary*. It is therefore only to the former class of minerals that the above-mentioned arguments are applicable. In the case of secondary minerals, the ratio of the unknown disintegration product to the uranium from which it has been formed should always be less than could be predicted from a knowledge of the age of the geological formation in which it occurs.

Without applying any quantitative test it is a matter worthy of note that several elements frequently occur associated with uranium in minerals; as examples may be mentioned lead, thorium, bismuth, and barium,

besides the gases hydrogen, argon, and helium, which are frequently found occluded in the mineral. The question which arises is therefore whether any of these elements are in any way connected with the radioactive transformations of uranium.

We may at once dispose of the existence of helium, for this gas has been shown to be produced from radium, and there is every reason to suppose that it has its origin in the radioactive atom, and is, in fact, the  $\alpha$  particle expelled during radioactive processes. Whether all  $\alpha$  particles consist of helium would seem in the light of recent investigations to be doubtful, and there is some foundation for the belief that it may in certain cases consist of hydrogen, which would account for the occurrence of this gas in uranium ores.

Whether or not there is any foundation for the belief expressed by Boltwood that argon may be expelled during the "rayless" transformations which we have seen to take place must for the present remain undecided.

In this connection may be mentioned the recent experiments of Sir William Ramsay,

who claims to have shown that this gas may, under suitable conditions, be produced from radium emanation. If the emanation was allowed to decay in a solution of copper sulphate, argon was produced instead of helium, and when dissolved in pure water it gave rise neither to helium nor argon, but to neon. Thus the transformation undergone by the emanation appears to depend on its environment, giving rise to different members of the helium group of elements under different circumstances. When the emanation was allowed to decay in a solution of copper sulphate the further remarkable result was found that at the conclusion of the experiment lithium could be detected spectroscopically in the solution, although the greatest care had previously been taken to free the solution from all traces of this element, and Sir William Ramsay concludes that copper has been decomposed into lithium under the influence of the radiations from the emanation. It is, however, as yet too early to discuss the exact significance of these experiments, and the confirmation and development of these

most important and remarkable discoveries will be awaited with the greatest interest.

With regard to the four elements—lead, thorium, bismuth, and barium—it has been quite recently shown by Boltwood that the latter three elements do not satisfy the quantitative conditions consequent on their being produced from uranium. With lead the case would, however, appear to be different, for the quantity of this element appears from a large number of analyses to bear a constant ratio to the quantity of uranium with which it is associated in primary minerals of the same age. On the other hand, those minerals which are known to be of greater antiquity contain a greater relative portion of lead than those known to have been more recently formed. These circumstances are interpreted by Boltwood as indicating that lead is the final disintegration product of uranium. Strong as this evidence may appear, there are grave difficulties in the way of accepting this conclusion as correct. The atomic weight of lead being 207 and that of uranium 240, there is a change of



only 33 units in the fourteen transformations necessary for the conversion of uranium into lead. In view of the fact that the atomic weight of radium emanation seems to be about 100, the above conclusion appears to be doubtful. A determination of the atomic weight of some of the other transformation products of uranium would therefore be desirable to remove this discrepancy in the evidence on the question.

## CHAPTER X

### RADIO-ACTIVITY AS A GENERAL PROPERTY OF MATTER

#### INTRODUCTION

THE study of the constituents of minerals, such as pitchblende, containing uranium, has shown that their radio-activity is due not only to the uranium which they contain, but to the whole series of substances exhibiting the property of emitting radiations to a greater or less extent. It will also be remembered that it has been shown that helium is produced by radioactive processes, and that other known chemical elements such as lead are probably also formed as a result of these transformations.

Thus it will be seen that radio-activity is not a property possessed by one form of matter only, and it becomes a question of interest not only to consider whether these

non-radioactive products of the radioactive elements are really completely devoid of the properties of the substances from which they are known to have been formed, but also to investigate the question whether the property of radio-activity is not at least in some measure shared by *all* known chemical elements.

Now radioactive transformations can be detected only by the radiations to which they give rise, so that if we are deprived of the means of detecting these radiations we are left powerless to investigate these subtle changes taking place in the atoms of the elements. That there may be many radioactive processes which leave us in ignorance of their existence is obvious from the following considerations.

It was shown when dealing with the  $\alpha$  radiation emitted by radioactive bodies that if the velocity of the positively charged particle projected from the active substance fell below a certain limit, the radiations ceased to produce any ionization in a gas or any action on a photographic plate or

phosphorescent screen placed in their path. When the  $\alpha$  particles had traversed a certain definite distance in air, called by Bragg their "range," their velocity was so far reduced that they ceased to produce any electrical, photographic or phosphorescent effect by which their presence could be detected. Hence, though it was certain that the particles were being projected with enormous velocities through the space beyond their range, their presence could not be detected by any known method. It follows, therefore, at once that if  $\alpha$  particles were to be projected from a radioactive atom with a velocity even slightly less than that with which the  $\alpha$  particles are emitted in the cases we have hitherto been considering, we might be wholly unconscious of their existence.

Whether such radiations do, in fact, arise from substances usually considered to be inactive must at present remain a matter merely of speculation; the question can be answered only by a refinement of our methods of detecting radio-activity or by some entirely new mode of investigating

these phenomena. In the meantime, therefore, it is useless to attempt to settle this question. There is, however, another allied question which is within the scope of legitimate inquiry, namely, whether or not any detectable radiations are emitted by ordinary chemical elements even though these radiations are obviously exceedingly feeble. The subject is one of extreme experimental difficulty, for not only are the effects under investigation exceedingly minute, but even though radiations should be detected proceeding from any ordinary chemical element, it is further necessary, before any conclusion can be drawn, to make sure that the apparent activity is actually due to the substance under investigation and not to some radioactive impurity present in quite small amount. As a matter of fact all observations in this direction are complicated by this uncertainty, and before considering the question raised above it will first be necessary to inquire to what extent the presence of small quantities of the radioactive element may vitiate the experiments.

## THE WIDE DISTRIBUTION OF RADIUM

Although radium is always found in very minute quantities, this element appears to be distributed very much more widely than was at first imagined. So far from being confined to the few minerals containing large quantities of uranium, its presence has been detected in a very large variety of substances. It should, however, be understood that it is only by employing the most refined methods of measurement that this wide-spread existence of radium has been demonstrated. It is therefore clear that except in the case of ores rich in uranium, the concentration of the radium is always quite extraordinarily small.

At the commencement of this work it was pointed out that the air has always the power of conducting electricity to a slight extent, and must be considered as being permanently in a state of ionization. This fact has led to the belief that radiations must be traversing the air in order to keep it in this condition, and numerous investigations have been undertaken to determine the nature and source of this radiation.

As a possible explanation of this state of affairs it occurred to Elster and Geitel that radium distributed in small quantities over the earth might at least partially account for the observed phenomena, and with a view to testing their hypothesis experiments were made to test whether radium emanation could be detected in the atmosphere.<sup>1</sup>

To do this, wires charged to a negative potential were exposed in the open air so as to collect on them the active deposit of radium or thorium should the emanation from either of these elements be present. On removing the wires after subjecting them to this treatment, it was always found that when tested by means of a very delicate gold-leaf electroscope they exhibited distinct radio-activity. Moreover, the activity so acquired was only of a temporary nature and was found to completely disappear in the course of a few hours. Measurements of the rate of decay of this activity showed that it was due to the active deposit from radium, which must therefore have been present in the atmosphere. In

<sup>1</sup> Elster and Geitel, *Phys. Zeitschr.*, II, 590, 1901.

some later experiments on the same subject it was shown by Elster and Geitel that the quantity of activity collected on a wire was much greater if, instead of exposing a charged wire in the open air, the experiment was carried out in a closed space such as a cavern, in which the emanation could collect in large quantity. There is therefore no doubt that small quantities of radium emanation exist in the atmosphere, and it seems probable that the supply is maintained from radium contained in the earth, for if the conditions are favourable to the accumulation of the emanation arising from the ground, as is the case in caves, the quantity of emanation was found to be greatly increased.

A very large number of experiments have been made by different observers to detect the presence of radium in different minerals and springs. It would, however, be out of place to attempt here to summarize the enormous number of data which have been accumulated on this point. It may suffice to mention that radium emanation has been detected by J. J. Thomson<sup>1</sup> in the ordinary tap water of

<sup>1</sup> J. J. Thomson, *Phil. Mag.*, IV, 356, 1902.



Cambridge, and that this gas was also found to be present in the water of New Haven by Bumstead and Wheeler.<sup>1</sup> The fact that radium does exist widely distributed in small quantities through the earth may be taken as established, and part at least of the ionization of the atmosphere must be ascribed to this cause. Whether or not this is the *only* source of ionization in the atmosphere is another question, and it is only by careful quantitative experiments that any further light can be thrown on the subject.

#### THE CONDUCTIVITY OF AIR CONTAINED IN CLOSED VESSELS

If we consider the conductivity of the air inside a closed space, there are several causes which may give rise to the ionization always found to exist. In the first place we have learnt that there is present in the air a certain quantity of radium emanation which will produce ionization inside a closed vessel ; for since this gas is distributed through the air, there will always be some contained in a vessel

<sup>1</sup> Bumstead and Wheeler, *Amer. Jour. Sci.*, XVII, 97, 1904.

when it is at first closed. The ionization due to this cause should, however, disappear in the course of a few weeks, by which time the activity of the emanation would have decayed virtually to nothing. This is not the case, for, on the contrary, the ionization is found to increase for some time after filling a vessel with pure air. There exists, however, the effect of the emanation *outside* the vessel which, on account of the active deposit which it produces, will give rise to ionization inside the vessel by reason of the penetrating radiations emitted by the latter. That this cause contributes to the effect under examination seems certain, but it is equally certain that it is insufficient to account for the whole ionization inside a closed vessel, and there are many reasons why this is so.

In the first place, as has been shown by Cooke,<sup>1</sup> it is impossible to reduce the ionization to less than about 30 per cent of its normal value by surrounding a metallic ionization vessel with screens of lead of very great thickness. Now this in itself is perhaps some-

<sup>1</sup> Cooke, *Phil. Mag.*, October, 1903.

what indecisive, but if we add to this the fact, which has been established independently by Strutt,<sup>1</sup> McLennan and Burton,<sup>2</sup> and Cooke,<sup>3</sup> that the value of the ionization inside a closed vessel depends on the material of which the vessel is constructed, it is obvious that there must be some radiation proceeding from the walls of the vessel itself; nor is this radiation due to the active deposit of radium formed on the walls of the vessel, for it cannot be removed by cleaning the surface of the enclosure before making experiments.

We are thus driven to the conclusion that all metals give out radiations capable of ionizing a gas, and there only remains the important question as to the origin of these rays. There are three possibilities to be considered. In the first place, the effect may be due to some kind of secondary radiation, produced by radiations from without falling on the vessel; in the second place, it may be due to some radiative impurity contained in the

<sup>1</sup> Strutt, *Nature*, February 19th, 1903.

<sup>2</sup> McLennan and Burton, *Nature*, February 26th, 1903.

<sup>3</sup> Cooke, *Nature*, April 2nd, 1903.

metal; and thirdly, the metals themselves may be spontaneously emitting rays, or, in other words, ordinary metals may be radioactive.

That the first two of these causes play an important part in producing the radiations from ordinary metals would appear to be the case from very abundant evidence, but what we are specially concerned with here is the all-important question whether after allowing for these disturbing influences there still remain any radiations emitted by ordinary metals, which must be attributed to some intrinsic property of the metals. We will proceed to review some of the most important information bearing on this point.

Although there is some positive evidence on this question, most of the reasons which have led to the belief that ordinary materials are radioactive are of a negative kind. If it is found impossible to detect any trace of any known radioactive substance present as an impurity in a body, then it is certain that if that body emits radiations, it must either itself be responsible for the production of rays, or there must be present some impurity hitherto

unknown which is giving rise to the radiations observed. As long as no such impurity has been separated from the body under investigation, it is only natural to believe that we are dealing with a real property of the substance. Even if this should prove to be so, it cannot be considered as definitely proved that the material which has been examined is radio-active in the true sense of the word; for it remains to be demonstrated that the energy set free, which is used to ionize the gas in the neighbourhood of the body, is derived from the energy of the atoms of the body, and not from some external source. In the absence of evidence to the contrary, it is frequently assumed that the power of ordinary metals to produce ionization is an indication of their radio-activity. If this assumption is legitimate, we shall see that at least a very large number of well-known substances exhibit to a greater or less extent the remarkable properties of elements such as radium. But it should be remembered that the evidence on this point is based on an assumption which is in any case doubtful.

## THE RADIATIONS EMITTED BY ORDINARY MATERIALS

Let us first consider the evidence as to the existence of some radioactive impurity in ordinary materials giving rise to the radiations which are known to exist. Regarded from an *a priori* standpoint it is not unlikely to expect that traces of radium and possibly of thorium might be found to exist in the ordinary metals of commerce, for these have been prepared from minerals which might contain small quantities of these radioactive elements. When we further consider that traces of radium have been detected in places where it would have been impossible to predict its existence, such as the water supply of Cambridge, it becomes of great importance to make sure whether radium exists in most metals, and if so, whether it is in sufficient quantity to account for their apparent radioactivity.

It may at once be stated that this question has been answered in the negative ; for although repeated searches have been made to detect the presence of emanation evolved by

the ordinary metals these efforts have all resulted in failure. Thus J. J. Thomson<sup>1</sup> was unable to detect the presence of radium in such metals as zinc, tin, lead, bismuth, and copper, and Righi,<sup>2</sup> Wood,<sup>3</sup> and Campbell<sup>4</sup> all failed to obtain any emanation from a variety of metals investigated by them.

These results dispose of the possibility of the existence of thorium, radium, or actinium in the metals tested. The possibility of the presence of uranium may be discarded, for it can easily be shown that if this metal were present in quantity sufficient to account for the radiations emitted, its presence should be demonstrable by chemical means, and this is not the case. In this connection it may be of interest to mention that Elster and Geitel<sup>5</sup> have quite recently detected very minute quantities of polonium in lead; but the amount was so exceedingly minute that it

<sup>1</sup> J. J. Thomson, *Brit. Assoc. Rep.*, 1904.

<sup>2</sup> Righi, *Cim.*, January, 1905, S. 53.

<sup>3</sup> A. Wood, *Phil. Mag.*, April, 1905.

<sup>4</sup> N. R. Campbell, *Phil. Mag.*, April, 1905, and February, 1906.

<sup>5</sup> Elster and Geitel, *Phys. Zeit.*, 1907.

required the greatest experimental skill to detect its presence at all, so that this discovery does not affect the general argument regarding the radiations emitted from ordinary metals.

Thus the negative evidence all goes in favour of the radio-activity of ordinary materials. Since we can discover no other source of the radiation emitted by the metals we conclude that the metals must themselves be radiating. Such an argument is, however, always weak, and is by itself somewhat unconvincing. Fortunately there is some, though it must be admitted rather scanty, evidence which tends to confirm this conclusion.

The problem of ascertaining whether there is any radiation emitted by ordinary metals when due allowance has been made for the presence of radioactive impurities, secondary radiation, and other disturbing causes, has been attacked by Mr. N. R. Campbell,<sup>1</sup> whose experiments seem to show that part of the ionization inside a closed vessel is due to rays emitted by the walls of the enclosure. The absorption of this radiation by air is comparable with that

<sup>1</sup> N. R. Campbell, *loc. cit.*



for the  $\alpha$  rays from radium, but differs somewhat for different materials. Assuming that this radiation is of the  $\alpha$  type, it is shown by Campbell that the "ranges" of the rays emitted by different materials can be measured. When this is done it is found that the ranges of the rays from each metal have a definite value, as would be expected if the metals were truly radioactive. But it must be remembered that the effects to be measured are extremely small and the experiments difficult to perform. It would therefore be rash to accept without further evidence the conclusion that radioactivity is a universal property of matter.

There are already several known radioactive elements constantly emitting radiations and thereby being transformed into other substances. It is interesting to contemplate that this most remarkable property may not be confined to these special forms of matter but may be shared to a greater or less extent by everything we see around us.

## CHAPTER XI

### THE MECHANISM OF RADIOACTIVE CHANGES

#### NATURE OF RADIOACTIVE CHANGES

HITHERTO only the properties of radioactive bodies and the nature of the changes going on during their transformation have been considered. Little or nothing has been said as to the mechanism by which these changes may be considered to be taking place. It has been shown that radioactive bodies are constantly giving out considerable quantities of energy during their transformations, and the question naturally arises as to the source of this energy. There are, broadly, two points of view from which the matter may be regarded. First, it may be supposed that the energy is derived from sources external to the radioactive bodies, and that by some property peculiar to these substances they are capable

of absorbing energy sufficient to account for their radio-activity. For this purpose it has been assumed that space is always being traversed by radiations unabsorbed by ordinary materials, but capable of being absorbed even by small quantities of the radioactive substances, and that consequently we are unaware of the existence of these radiations except by the intervention of radioactive bodies. Ingenious as this hypothesis appears, there is no evidence to support it; for though it seems certain that there is always traversing space a penetrating type of radiation, yet, as far as can be tested by experiment, this radiation has nothing whatever to do with the phenomena of radio-activity. In the first place, the energy which might be supposed to be communicated to a radioactive body in this way is too small to account for the heat emitted by it; and secondly, it is impossible to influence the activity, say of radium, however great be the precautions observed to screen it from external radiation. For these and other reasons it is difficult to reconcile the view that radio-activity is due to the

absorption of radiant energy derived from some source external to the radioactive body, and it seems more rational to look to the radiating substance itself to discover the source of the energy which it is continually emitting.

Now it must be remembered that during radioactive processes, accompanying the evolution of heat there is a continual change of one kind of material into another, and it would be not unnatural, at first sight, to suppose that the changes involved are chemical in nature, for these too consist of material transformations accompanied by absorption or evolution of heat. A closer examination of the matter, however, shows this view to be quite untenable, for not only is the amount of energy set free by a minute quantity of radioactive material during its transformation enormous compared with the energy evolved by the same quantity of matter undergoing any known chemical change, but, as we have seen, the rate at which radioactive substances are transformed is unaffected by greatly changing the conditions to which they are subjected.

Thus, for example, the radiation emitted by radium is the same at the temperature of liquid air as at a white heat. This is quite inconsistent with the view that the changes involved in the transformation of radium are in any way similar to those taking place during chemical change.

There is yet further evidence which tends to differentiate the radioactive changes from chemical reactions. It has been seen that the activity of the radioactive elements is, at least, within wide limits, independent of their environment; for they may be subjected to a variety of physical conditions without suffering any alteration. But this is not all, for they may be chemically transformed without being in the least changed so far as their radio-activity is concerned. Thus different salts of the same radioactive element have the same activity provided that they contain the same quantity of that element: for example, uranium oxide has the same activity as uranium nitrate if the quantities of these substances are so chosen as to contain equal quantities of uranium, and the same rule

holds good if the activity of metallic uranium is compared with that of any of its salts. The same is true of thorium and of the salts of radium. It is true that this latter element has never yet been obtained in the free state, but there is no reason to believe that it would behave differently from uranium and thorium in this respect.

All these facts have led to the belief that radioactive transformations, unlike chemical changes, do not depend upon the interactions of the atoms of which they are composed, but are the manifestation of changes going on within the atom, and are thus of a more subtle nature than the reactions of chemistry. In order, therefore, to understand the views which are now held with regard to radioactivity, it is necessary to consider what information has been gathered with regard to the constitution of atoms in general, and to see how far the knowledge thus gained is of assistance in explaining the phenomena of radio-activity.

## THE CONSTITUTION OF THE ATOM

The science of chemistry has demonstrated that all forms of matter can be built up by suitable combinations of some seventy simple substances known as elements. The atoms of which these elements are made up are indivisible by any chemical reaction, and are therefore the simplest forms of matter with which chemistry has to deal. It has long been felt that a great simplification would be introduced if all the elements could be shown to consist of atoms of the same kind, by the combinations of which in different proportions the elements might be supposed to be built up. The conception that all matter might thus be ultimately reduced to one primordial form is no new one, and speculations to explain the constitution of matter on these lines have at all times been plentiful.

The first truly scientific attempt to unify matter in this manner was made in the year 1815, when Prout put forward the hypothesis that all matter was ultimately composed of

hydrogen, the lightest known element. In support of his view Prout showed that the atomic weights of the commoner elements were multiples of that of hydrogen, so that by the condensation of the atoms of the latter with each other the various elements might be evolved. This important suggestion was in agreement with the facts known at the time it was propounded, but subsequent and more accurate determinations of the atomic weights of the elements, notably those of chlorine, potassium, sodium, and silver, by Stas, have shown that the hypothesis cannot be accepted in the form in which it was put forward. Unsuccessful attempts have since been made to modify the hypothesis in such a way as to render it consistent with the results of experiments. Thus it was suggested that by taking some submultiple of the atomic weight of hydrogen as the ultimate constituent of all atoms the hypothesis in this modified form might be retained. One-half, one-quarter, and even smaller fractions of the hydrogen atom were selected as the ultimate unit from which the elements were built up.



But these speculations were unsupported by any evidence and incapable of being experimentally tested, so that the hypothesis for a time fell into disrepute. It is, however, now realized that, taking the atomic weight of oxygen as sixteen, a larger proportion of the atomic weights of the other elements than can be ascribed to chance are very nearly whole numbers. The precise significance of this is unknown, but the fact is certainly remarkable.

Of recent years the view that atoms of the elements are made up by the accumulation into groups of different numbers of some primary unit of small size has again come into favour. A multitude of evidence derived from a study of electrical and optical phenomena has led to the conclusion that there must be within all atoms certain minute systems in constant vibration, and that these systems are made up of precisely the same corpuscles or electrons which as we have seen constitute the cathode rays and  $\beta$  rays from radioactive substances. These minute particles have been seen to carry a negative

charge, and when moving with sufficiently high velocities possess mechanical inertia in virtue of their rapid motion. In view of these facts and the evidence which leads to the belief that within the atom there must be a greater or less number of corpuscles in rapid motion, the suggestion has been put forward that the inertia of matter, as we know it, may be explained in terms of these moving corpuscles. Although the attempt to thus explain the properties of matter in terms of electricity in motion has not been altogether successful, the idea has greatly increased our knowledge of the constitution of atoms and, what is of greater importance from our present point of view, has shed considerable light upon the possible causes of the phenomena of radio-activity which we have already seen to be the manifestation of changes going on within the atoms.

Now, it will be remembered that when dealing with the properties of the radiations from radioactive substances, it was shown that as a result of the experiments of Kaufmann with  $\beta$  rays moving with different velocities, the

corpuscle could be considered as possessing inertia purely electrical in its origin. This is true of the negatively charged radiations given out from radioactive bodies; but the same has not yet been demonstrated for the  $\alpha$  rays which carry with them a positive charge, and consist of particles whose mass is comparable with that of the hydrogen atom. This fact has given rise to the belief that there is some fundamental difference between positive and negative electricity, for whereas it is possible to isolate a negatively charged corpuscle with purely electrical inertia, experiment has failed to reveal an analogous corpuscle with a positive charge. The state of our knowledge regarding the nature of a positive electricity therefore falls behind that relating to negative electricity, and the investigation of the nature of the former constitutes one of the most important problems of physics, without an understanding of which the constitution of the atom must remain at best indefinite. If, however, we disregard this outstanding difficulty, we shall see that important results follow from the

view that within every atom there are a number of negatively charged corpuscles always in rapid motion.

It is clear that if this conception of the atom is to be adopted there must be besides the negatively charged corpuscles an equal charge of positive electricity associated with the atom. For not only would the atom be unstable without this, but since the atoms are themselves electrically neutral, it is obviously necessary to postulate the existence of a positive charge to neutralize the free negative charges supposed to exist within the atom.

There are many hypotheses which have been put forward as to the distribution of the positive charge in the atoms, but the most suggestive of these is that proposed by J. J. Thomson. According to this view the atom consists of a number of corpuscles in rapid motion within a sphere of uniformly distributed positive electricity. Although there is no particular reason for adopting this precise distribution of the positive electricity, this assumption has the great ad-

vantage that it lends itself to comparatively simple mathematical treatment. According to the scheme proposed by J. J. Thomson, negative corpuscles are supposed to be in rapid rotation in orbits within the sphere of positive electricity. Under these circumstances the corpuscles form themselves into concentric rings, and it can be shown that there are only quite definite configurations of the corpuscles within the atom which are stable. Thus it can be shown that with less than five corpuscles within the atom these corpuscles would arrange themselves in a single ring. On adding a sixth, a discontinuity of arrangement would occur, and the stable system would now consist of a ring of five corpuscles with one at the centre. On further increasing the number of corpuscles the two-ring system would persist until there were fifteen corpuscles within the atom, when a three-ring system would be formed, and so on for greater numbers.

This, then, is in general principle the conception of the constitution of the atoms which we are considering, and in support of this view

J. J. Thomson has shown that many of the facts of chemistry can be explained by considering the atoms of the various elements as made up of such systems containing different numbers of corpuscles. But with these matters we are not here concerned. Suffice it to say that there is considerable evidence to indicate that some such constitution of the atom has much to recommend it, both from physical and chemical evidence.

Let us consider what light this theory throws on the facts of radio-activity. It has already been seen that a system of rotating corpuscles arrange themselves into quite definite configurations. But it can be shown that if the velocity of rotation is increased above a certain critical value, other configurations may suddenly become stable. Suppose, then, that we have a system of corpuscles rotating with velocities above this critical value, certain configurations will be stable which could not exist if the velocity were reduced below the critical velocity. Now it can be demonstrated that such a system will continually radiate out energy, though

possibly at a very slow rate, and this energy will be derived from the energy of rotation of the corpuscles. The velocity of the corpuscles will thus be slowly reduced, and must inevitably reach the critical value below which they are no longer in stable equilibrium. A complete rearrangement of the corpuscles in the atom will suddenly occur, and during the violent disturbance which must thereby be caused, certain portions of the atom may break free which manifest themselves as radiations from the atom. This is what may be conceived to be taking place with the radioactive elements. If such explosions of the atoms occur frequently we have a strongly radioactive element. If they occur less frequently, or not at all, we have a feebly radioactive or non-radioactive substance, as the case may be.

It is difficult to say how far such a conception of radioactive changes actually represents the phenomena which are taking place within the radioactive substances, but the theory we have briefly considered above affords some mental picture of the violent

changes which must take place during the disintegration of a radioactive atom. It is remarkable that a theory of matter which has been devised to explain the physical and chemical behaviour of atoms in general should be capable of interpreting the recently discovered property of radio-activity possessed to an appreciable extent by certain forms of matter only.



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